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Revisiting and modeling the magnetism of hole-doped CuO_2 spin chains in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$

J. Schnack^{a,*}, R. Klingeler^b, V. Kataev^b, B. Büchner^b

^aUniversity of Osnabrück, Dept. of Physics, D-49069 Osnabrück, Germany

^bInstitute for Solid State and Materials Research IFW Dresden, P.O. Box 270116, D-01171 Dresden, Germany

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Abstract

Magnetization measurements of $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ with $0 \leq x \leq 12$ in magnetic fields up to 16 T show that the low-temperature magnetic response of the CuO_2 spin chains changes strongly upon Ca doping. For $x = 0$ quantum statistical simulations yield that the temperature and field dependence of the magnetization can be well described by an effective Heisenberg model in which the ground state configuration is composed of spin dimers, trimers, and monomers. For $x > 0$ a constant contribution to the low-temperature magnetic susceptibility is observed which cannot be explained in terms of simple chain models. Alternative scenarios are outlined.

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1. $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$

The low-temperature magnetic response of $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ is dominated by the CuO_2 chain subsystem, whereas the ladder subsystem is magnetically silent due to a large spin gap. The magnetic susceptibility, shown in Fig. 1, exhibits two pronounced features, a clear maximum at about 75 K and a Curie-like divergence at $T = 0$. The maximum has been attributed to the existence of antiferromagnetically coupled dimers on the chains, which are supposed to host nearly six Zhang-Rice singlets (“holes”) per 10 Cu sites. The $T = 0$ divergence was attributed to free Cu spins residing somewhere in the crystal structure. In a recent investigation [1] it could be shown by means of EPR measurements that these impurities are located on the chains since they experience the same chemical environment as the Cu ions bound in dimers. Taken this into account a quantum statistical simulation – as outlined in Ref. [2] – was performed in order to treat both dimers and monomers and other possible substructures on a common footing. The model is similar to a simple Born-Oppenheimer description where the electronic Hamiltonian (here spin Hamiltonian) depends parametrically on the positions of the clas-

sical nuclei (here hole positions). Each configuration \mathbf{c} of holes and spins defines a Hilbert space which is orthogonal to all Hilbert spaces arising from different configurations. The Hamilton operator $\tilde{H}(\mathbf{c})$ of a certain configuration \mathbf{c} is of Heisenberg type, i. e.

$$\tilde{H} = \sum_{\mathbf{c}} \left(\tilde{H}(\mathbf{c}) + V(\mathbf{c}) \right) \quad (1)$$

$$\tilde{H}(\mathbf{c}) = - \sum_{u < v} J_{uv}(\mathbf{c}) \mathbf{s}(u) \cdot \mathbf{s}(v) \quad (2)$$

$$V(\mathbf{c}) = \frac{e^2}{4\pi\epsilon_0 \epsilon_r r_0} \frac{1}{2} \sum_{u \neq v} \frac{1}{|u - v|} \quad (3)$$

$J_{uv}(\mathbf{c})$ are the respective exchange parameters which depend on the configuration of holes. For the theoretical results four exchange parameters are used. The strongest and antiferromagnetic exchange $J = -134$ K is across one hole. The exchange across two holes $J_{\parallel} = 15$ K is ferromagnetic as is the exchange $J_{NN} = 100$ K of neighboring spins. We also apply a coupling across one spin which is weaker than J and taken to be $J_s = -34$ K. The following statements are robust for reasonable variations of J_{\parallel} , J_{NN} , and J_s . Periodic boundary conditions are applied for the following calculations.

Two very good fits are shown in Fig. 1 (lines) together with the experimental data points (crosses). The fits yield

* Corresponding author.

Email address: jschnack@uos.de (J. Schnack).

that it is equally well possible that the chains contain monomers as well as that they contain trimers. The first case is very similar to the one found using density functional theory calculations [3,4].

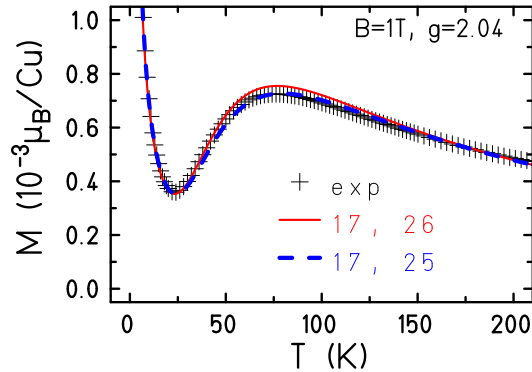


Fig. 1. Magnetization versus temperature for $B = 1$ T: Experimental data (B parallel to the c -axis) are given by crosses. The results of a complete numerical diagonalization are depicted by a solid curve for $N_s = 17$ and $N_h = 26$ as well as by a dashed curve for $N_s = 17$ and $N_h = 25$.

2. $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$

For the calcium doped compounds $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ the picture is much more involved. Whereas $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ exhibits charge order on the chains due to the combined presence of long range Coulomb interaction like in $\text{Na}_{1+x}\text{CuO}_2$ [5] and commensurate doping level, the Ca-doping results in a decreasing stability of charge order [6,7,8]. In addition, the dimer spin gap is strongly affected by the doping level, and at low temperatures a finite susceptibility, which is not Curie-like, is observed [9]. This susceptibility increases with the Ca content. Figure 2 shows the temperature dependence of the magnetization for various Ca-doped compounds for $B = 1$ T.

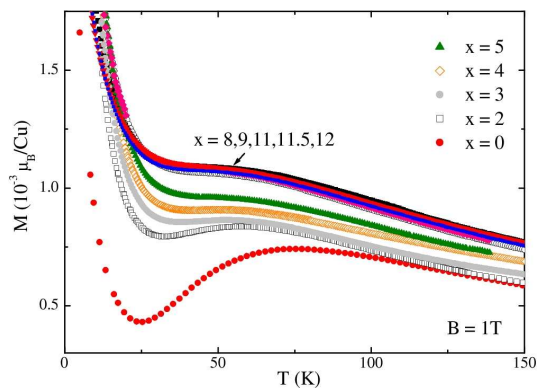


Fig. 2. Magnetization of $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ with $0 \leq x \leq 12$ in a magnetic field of $B = 1$ T parallel to the c -axis.

It is known that application of chemical pressure through Ca-doping leads to a hole transfer from the chains to the ladders in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ [10,7]. The estimated maximal reduction of the number of holes on the chains is

one hole out of six per formula unit. In addition, it is usually assumed that the increased presence of holes on the ladders does not significantly reduce the ladder's spin gap. Therefore, the low-temperature low-field magnetic response should still origin from the chain subsystem only.

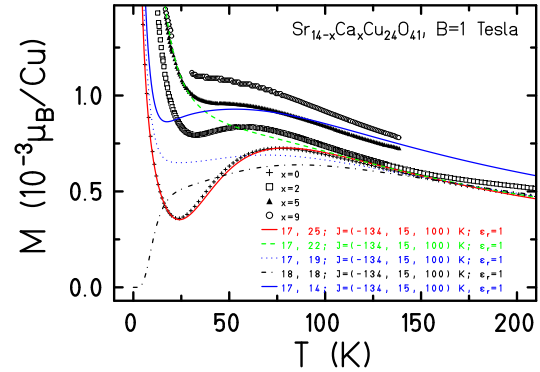


Fig. 3. Magnetization of model spin-hole chains for various numbers of spins and holes (curves). Experimental data are given by symbols.

In the context of the quantum statistical model, Eqs. (1)–(3), we investigate a huge variety of doping levels numerically. The number of spins is $N_s = 16, 17$, or 18 , whereas the number of holes is varied in the range $26 \geq N_h \geq 10$. Figure 3 shows a small selection of resulting magnetization curves. A striking feature of all curves – not only those which are shown – is that for $N_h \geq N_s$ the high temperature part at $T = 100 \dots 150$ K does not increase with decreasing number of holes. Only for hole levels significantly less than 50 % the chains show a magnetization comparable to the experiment.

The discrepancy between experimental and theoretical results has the following consequences. Antiferromagnetic spin chains of finite length (50 % holes) cannot account for the observed behavior. In general, hole levels above 50 % cannot produce the observed high-temperature magnetization. In addition both scenarios cannot explain the low-temperature high-field magnetization. Thermal and field induced dynamics of the holes might be a solution. In addition, the assumption of a gapped spectrum of hole-doped ladders needs to be justified. On the theory side the assumption of hole localization should be relaxed, i.e. the system should be investigated in the context of a generalized Hubbard model.

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