Revisiting the chain magnetism in $Sr_{14}Cu_{24}O_{41}$: Experimental and numerical results

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We study the magnetism of the hole doped CuO_2 spin chains in $Sr_{14}Cu_{24}O_{41}$ by measuring the Electron Spin Resonance (ESR) and the static magnetization M in applied magnetic fields up to 14 T. In this compound, the dimerized ground state and the charge order in the chains are well established. Our experimental data suggest that at low temperatures the Curie-like increase of M as well as the occurrence of the related ESR signal are due to a small amount of paramagnetic centers which are not extrinsic defects but rather unpaired Cu spins in the chain. These observations qualitatively confirm recent *ab initio* calculations of the ground state properties of the CuO_2 chains in $Sr_{14}Cu_{24}O_{41}$. Our complementary 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.

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I. INTRODUCTION

Hole doped antiferromagnets with an inhomogeneous ground state have attracted much attention in recent years. A prominent example is provided by high temperature (high-T_c) superconductors in which low dimensionality, $s = 1/2$ quantum magnetism, and charge degrees of freedom are intimately related. In particular, a nonuniform distribution of charges in the Cu-O planes of the high-T_c layered cuprates seems to play an important role in establishing the superconductivity.[1] The quasi one-dimensional (1D) edge-sharing CuO_2 chain compounds such as $(Sr, Ca, La)_{14}Cu_{24}O_{41}$, $Na_{1+x}CuO_2$, or $Ca_{2+x}Y_{2-x}Cu_5O_{10}$ constitute another class of cuprates with charge inhomogeneities.[2,3,4,5] The interplay of spin and charge degrees of freedom in one dimension, as is evidenced by both theoretical and experimental results, is even more pronounced as compared to the layered 2D situation, yielding unusual ground states and novel excitations both in the spin and charge sectors.

In this work we focus on the low-temperature magnetic properties of the so-called "telephone number" compound $Sr_{14}Cu_{24}O_{41}$, where a composite structure with Cu_2O_3 spin ladders and CuO_2 spin chains is realized. Magnetically, the ladders do not contribute significantly at low temperature due to a large spin gap of $\Delta \sim 380\,\mathrm{K}$.[6,7,8] A remarkable property of this compound is self-doping, i.e. the material is intrinsically hole doped with 25% of holes per Cu site. As most of the holes are localized in the chains their concentration in this subunit is very high, close to 60% per Cu in the chain formula unit (c.f.u.). This influences strongly the magnetism of the spin chain resulting in an inhomogeneous ground state. Two Cu spins which are separated by a hole site couple antiferromagnetically (AF). If they are surrounded by two localized holes at both sides they form almost independent dimers. Such a dimerized nonmagnetic state in the chain subsystem of $Sr_{14}Cu_{24}O_{41}$ is experimentally well established.[9,10,11,12] The occurrence of charge order, which is an important ingredient of the dimer scenario in $Sr_{14}Cu_{24}O_{41}$, is supported e.g. by NMR and ESR data.[13,14] In a very recent numerical study[15] Gelle and Lepetit (GL) highlight that structural modulations due to the pseudoperiodicity of the chains and the ladders might be crucial for the electron localization and for the occurrence of the dimer state. Remarkably, these calculations suggest that the ground state is not perfectly dimerized even in the limiting case where all holes are located in the chains. Ideally, in that case, where six out of the ten sites in the chain unit cell are holes, the remaining four spins could be coupled to two AF dimers with no free spins left. In contrast to that, GL arrive at the surprising result that it is energetically favorable if a few spins are left unpaired. Then the stable configuration corresponds to 1.7 dimers and 0.5 free spins per c.f.u., respectively. Therefore, the occurrence of a Curie contribution to the magnetic susceptibility of $Sr_{14}Cu_{24}O_{41}$ at low temperatures, which has originally been attributed to the free spins, might be an intrinsic property of the hole doped Heisenberg Cu-O spin chain due to the distortion by structural modulations.

The aim of our paper is thus twofold. First, we

show experimental magnetization and electron spin resonance (ESR) data which give strong indications that a small amount of free spins, that gives rise to a finite magnetization and to an ESR signal at low temperatures, is not extrinsic but mainly reside in the chain subunit of $Sr_{14}Cu_{24}O_{41}$. This finding *qualitatively* supports the model calculations by GL in Ref. 15. Secondly, we present our own quantum statistical simulations which rely on the numerical diagonalization of an effective Heisenberg Hamiltonian.[16,17] This Hamiltonian, which depends parametrically on hole positions and on the screened Coulomb interaction between them, also yields an inhomogeneous ground state configuration of spins and holes. It turns out that the ground state is dominantly built of the aforementioned dimers, but also contains weakly coupled monomers and/or trimers which not only leads to the Curie-like behavior of the magnetization at low temperatures, but also yields a nearly perfect *quantitative* description of the low-temperature magnetic response of the Cu spin chain. The very good agreement also suggests that other arrangements of spins and holes are well gaped from the ground state configuration.

II. EXPERIMENTAL RESULTS

In this section we present ESR and magnetization measurements on a high quality single crystal of $Sr_{14}Cu_{24}O_{41}$. For ESR experiments an X-Band Bruker ESR spectrometer has been used, which operates at a frequency of 9.5 GHz. The magnetization data have been collected with a homemade vibrating sample magnetometer (VSM) in external magnetic fields up to 14 T and with a SQUID magnetometer in a field of 1 T. The single crystal grown by the floating zone technique[18] has been previously thoroughly characterized by measuring the magnetization, the thermal expansion,[19] the electrical and thermal transport,[8,20] the inelastic neutron scattering cross section,[11], and the ESR response.[14]

In order to recall the magnetic properties of $Sr_{14}Cu_{24}O_{41}$ we show in Fig. 1(a) the temperature dependence of the magnetization $M(T)$ of our single crystal in a magnetic field of $B = 1$ T applied parallel to the crystallographic c-axis (chain direction). The $M(T)$ dependence can be very well fitted with Eq. (1) comprising two terms, the magnetization M_{dimer} of AF coupled dimers in a concentration N_D with an exchange constant $J/k_B = -134$ K, and the Curie magnetization M_{Curie} owing to a small concentration N_S of free spins (see, e.g. Ref. 14)

$$M(T) = M_{dimer} + M_{Curie} = \frac{N_A g^2 \mu_B^2 B}{24 k_B}\left[\frac{2N_D}{T[3+\exp(-J/k_BT)]} + \frac{N_S}{4T}\right] . \quad (1)$$

The very good agreement of the experimental $M(T)$ curve with Eq. (1) give evidence of the dimerized ground

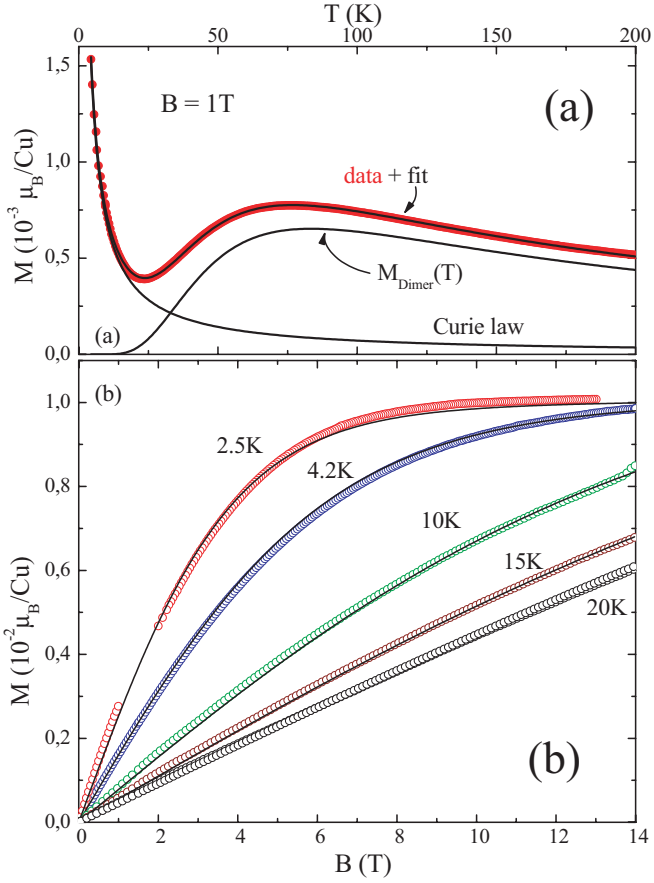

FIG. 1: (color online) Magnetization of $Sr_{14}Cu_{24}O_{41}$ versus temperature for a constant magnetic field of $B = 1$ T (a) and versus applied magnetic field for 2.5 K$\leq T \leq$ 20 K (b). B was applied along the c-axis. Lines are fits to the data (see text).

state which in addition has been proven for our sample by inelastic neutron scattering and thermal expansion measurements.[11,19] In Fig. 1(b) we present the magnetization versus applied magnetic field, $M(B)$, at temperatures of 20 K, 15 K, 10 K, 4.2 K, and 2.5 K, respectively. At all temperatures the dependence of M on B can be nicely fitted as a sum of the temperature independent contribution $\chi_0 \cdot B$ owing to diamagnetism and Van-Vleck paramagnetism of the Cu ions and a Brillouin function $B_{s=\frac{1}{2}}$ describing the magnetization of free $s = 1/2$ spins with a concentration N_S

$$M(B) = \chi_0 \cdot B + \frac{1}{2} N_S g_c \mu_B \cdot B_{s=\frac{1}{2}}\left(\frac{g\mu_B(B+\lambda M)}{2k_BT}\right) . \quad (2)$$

With a g factor of $g_c = 2.04$ (see below) one obtains $\chi_0 \approx 1 \cdot 10^{-5}$ emu/Mol Cu, the mean field parameter $\lambda = 0$ and $N_S \simeq 0.01$/Cu. We note that the magnetization at $T = 2.5$ K saturates in a field of $B = 14$ T at a very small value of about $1 \cdot 10^{-2}\,\mu_B$ /Cu justifying that the magnetic response at low temperature is caused by

about 1% of free $s = 1/2$ spins while there is no response from the remaining 99% of the spins. Moreover, we emphasize that the determination of N_S from our high field magnetization data is much more straightforward and accurate as compared to the conventional analysis of the low temperature contribution to $M(T)$ using the M_{Curie} term in Eq. (1). Therefore, precise knowledge of N_S from the analysis of $M(B)$ curves allows for an accurate determination of the number of dimers N_D. Substituting $N_S = 0.01$/Cu in the M_{Curie} term in Eq. (1) one obtains from the fit to the experimental $M(T)$ dependence $N_D = 0.0738$/Cu. Thus the number of magnetic Cu sites amounts to $N_S + 2N_D \approx 3.78$/f.u. in our $Sr_{14}Cu_{24}O_{41}$ single crystal which differs from a previous less precise estimate that employed the analysis of $M(T)$ curves of polycrystalline samples.[21]

We note that the concentrations of dimers and free spins, N_D and N_S, respectively, are significant observables in the recent *ab initio* calculations of the low energy electronic properties of the Cu-O chain in $Sr_{14}Cu_{24}O_{41}$ by GL.[15] Therefore, the accurate determination of N_D and N_S from our magnetization data provides an important check for the relevance of the GL model with respect to the low energy physics of $Sr_{14}Cu_{24}O_{41}$. Comparison of our data with the numerical results shows that the number of dimers N_D found in our analysis roughly agrees with the calculations of GL for the case that all holes are localized in the chains, $N_D^{GL} = 0.0708$/Cu . The number of free spins N_S, however, is smaller than theoretically predicted $N_S^{GL} = 0.021$/Cu by a factor of two. This is surprising because the experimental value N_S comprises not only the response owing to the imperfect dimerization of the spin chain, but also should include the response of paramagnetic defects which may occur in real crystals. Thus, theoretical calculations should yield a smaller number of free spins than experimental data. Besides the quantitative discrepancy this comparison suggests that, at least in the framework of the GL model, the free spins in a concentration $N_S \simeq 0.01$/Cu found in our $Sr_{14}Cu_{24}O_{41}$ single crystal are *not extrinsic* paramagnetic defects but mainly reside in the spin chain. This suggestion is corroborated with the analysis of the anisotropy of the ESR signal and of the magnetization.

Detailed studies of the ESR response from the spin chains in the family of the "telephone number" compounds have been published in Refs. 14,22. Here we present the ESR data of the $Sr_{14}Cu_{24}O_{41}$ single crystal in the low temperature regime. Below 20 K the ESR signal is mainly due to the free spins, which also contribute to the Curie-like increase of the magnetization, compare Fig. 1(a). At higher temperatures the ESR response is dominated by the dimer spins which are thermally activated. The evolution of the g factor of the ESR signal in the crossover regime around 20 K is shown in Fig. 2. The g factor of the Cu spins in a dimer is anisotropic with the values $g_c = 2.045$ for the magnetic field $\vec{B}$ parallel to the chain direction (c-axis) and $g_b = 2.284$ for the magnetic field $\vec{B}$ perpendicular to the plane of the chains (b-axis),

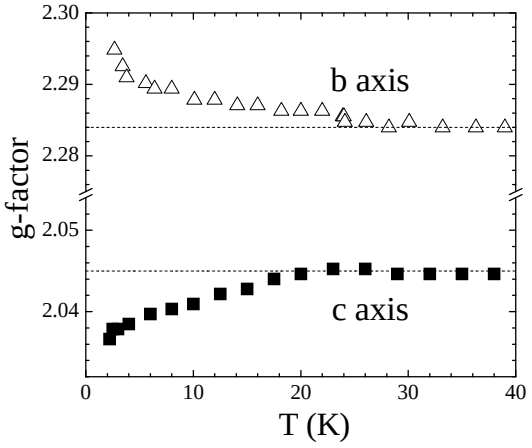

FIG. 2: Temperature dependence of the g-factor along c- and b-axis, respectively, as obtained from ESR data. Dashed lines extrapolate g of the dimers. Below ∼20 K, the response is due to small deviations from the perfectly dimerized state.

respectively. These g factors are temperature independent. The anisotropy of the g tensor is determined by the crystal field splitting of the Cu orbital state of e_g and t_{2g} symmetry

$$g_c = 2 - \frac{8 \cdot \lambda}{\Delta_{yz,zx}}, g_b = 2 - \frac{2 \cdot \lambda}{\Delta_{xy}} \ . \qquad (3)$$

Here $\lambda = -0.1$ eV is the spin-orbit coupling constant and $\Delta_{yz,zx}$ and Δ_{xy} are the energy differences between relevant orbital states.[23] The anisotropy of the g tensor above $\sim$ 20 K is temperature independent and is typical for a Cu ion in an approximately square planar coordination of the oxygen ligands,[14,23] i.e. the coordination occurring in the chain of $Sr_{14}Cu_{24}O_{41}$. Remarkably, below 20 K the g factor for both orientations changes only slightly giving strong indication that the ESR response at low Ts comes from a paramagnetic Cu ion having a very similar environment to that of the chain Cu ions. A small change in the anisotropy of the g factors can be explained by a slightly different charge distribution around a free Cu spin which has in the chain two neighboring holes, both to the left and to the right hand side, whereas a Cu spin in a dimer has only one neighboring hole on the one side and two holes on the other side, compare also Fig. 5.

The fact that the free spins exhibit the similar anisotropy as the dimerized ones is confirmed by our magnetization data. Our analysis of $M(T)$ shows that the ratio of the Curie constants which describe the response of the free spins along b- and c-axis amounts to $C_b/C_c = g_b^2/g_c^2 \simeq 1.25$. This ratio agrees well with our ESR data. This fact is illustrated by Fig. 3 in which M/g^2 is shown for B parallel to the b-axis and the c-axis, respectively. After being corrected for the anisotropy of the g factor the magnetization data for two orientations coincide almost perfectly in the whole temperature range. Note that in our analysis we have also considered the

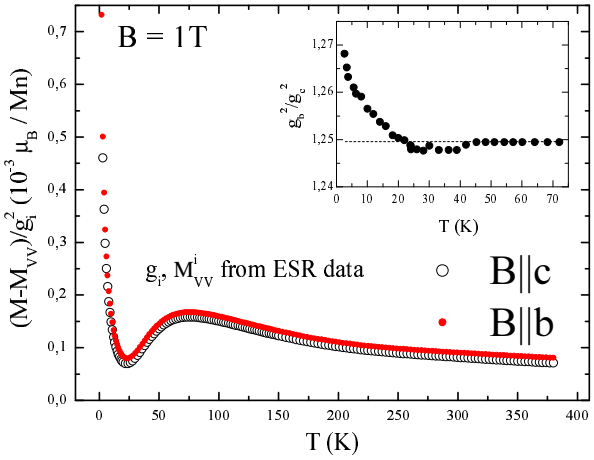

FIG. 3: (color online) Magnetisation divided by the g-factor and corrected by anisotropic Van-Vleck magnetism along the respective axes of $Sr_{14}Cu_{24}O_{41}$ for a constant magnetic field $B = 1$ T versus temperature.

anisotropy of the Van-Vleck magnetism which is related to the anisotropy of the g factors as[23]

$$\chi_c^{VV} = \frac{8 \cdot \mu_B^2}{\Delta_{yz,zx}} = (2 - g_c)\frac{\mu_B^2}{\lambda} \tag{4}$$

$$\chi_b^{VV} = \frac{2 \cdot \mu_B^2}{\Delta_{xy}} = (2 - g_b)\frac{\mu_B^2}{\lambda} \ . \tag{5}$$

To summarize these results, our experimental data strongly suggest that the free spins which cause the Curie-like response in $Sr_{14}Cu_{24}O_{41}$ are not extrinsic paramagnetic defects but reside in a not perfectly dimerized chain. Thus our experimental study qualitatively confirms the results of GL in Ref. 15, which predict the occurrence of such free spins in the hole doped chain as a result of structural modulations. As noted above, the theoretical results suggest, however, a larger Curie-like response than observed experimentally. In addition, while GL assume slightly more than 60% of holes in the chains of $Sr_{14}Cu_{24}O_{41}$, recent NEXAFS experiments imply that there are less holes in the chains,[24] i.e. there is a larger number of magnetically active spins in the chains.

To resolve this contradiction we perform a numerical study to be presented in the next section which gives evidence for trimers in spin chains with less than 60% hole doping. Trimers consist of three next-nearest neighbor spins with a hole between each spin, which are confined by two holes at each side, compare Fig. 5. The presence of trimers might lift the discrepancy between the calculated number of paramagnetic defects in the dimerized state and the smaller experimental value of N_S. For small external fields, trimers respond similarly to free spins. The number of magnetically active spins involved in this response is, however, three times larger. The first non-trivial excitation of trimers occurs at $1.5J \sim 200\,\mathrm{K}$, which can not be observed in our $M(B)$ data at 2.5 K because this is far above the accessible field range. On the other hand, our $M(T)$ data do not rule out this scenario because at higher temperatures there are indeed additional contributions to M which are usually ascribed to the magnetism of the ladders and to the melting of the charge order. Our numerical analysis in section III shows, that trimers (instead of or in addition to single spins) perfectly explain our experimental data. In contrast, the presence of quadrumers, which exhibit the spin gap of $0.659J \sim 90\,\mathrm{K}$ is excluded by our $M(T)$ data shown in Fig. 1(a). Our data also rule out that the Curie-like response is due to nearest neighbor dimers, trimers, etc., which are coupled ferromagnetically.

III. EXACT DIAGONALIZATION STUDIES

The dynamics of a hole-doped chain system constitutes a real challenge for theoretical investigations. Especially the evaluation of thermodynamic quantities both as function of temperature and magnetic field is prohibitively complicated even for moderate system sizes. In the following we are using an effective spin Hamiltonian which depends parametrically on hole positions.[17] This ansatz is similar to a simple Born-Oppenheimer description where the electronic Hamiltonian (here spin Hamiltonian) depends parametrically on the positions of the classical nuclei (here hole positions). Each configuration $\vec{c}$ of holes and spins defines a Hilbert space which is orthogonal to all Hilbert spaces arising from different configurations. The Hamilton operator $\underset{\sim}{H}(\vec{c})$ of a certain configuration $\vec{c}$ is of Heisenberg type, i. e.

$$\underset{\sim}{H} = \sum_{\vec{c}} \left(\underset{\sim}{H}(\vec{c}) + V(\vec{c}) \right) \tag{6}$$

$$\underset{\sim}{H}(\vec{c}) = -\sum_{u<v} J_{uv}(\vec{c})\ \underset{\sim}{\vec{s}}(u) \cdot \underset{\sim}{\vec{s}}(v) \tag{7}$$

$$V(\vec{c}) = \frac{e^2}{4\pi\epsilon_0\,\epsilon_r\,r_0}\frac{1}{2}\sum_{u\neq v}\frac{1}{|u-v|} \ . \tag{8}$$

$J_{uv}(\vec{c})$ are the respective exchange parameters which depend on the configuration of holes. $J < 0$ describes antiferromagnetic coupling, $J > 0$ ferromagnetic coupling. For the theoretical results presented in this article three exchange parameters are used, see Fig. 4. The strongest and antiferromagnetic exchange $J = -134$ K is across one hole. The exchange accross two holes $J_{\|} = 14$ K is ferromagnetic as is the exchange $J_{NN} = 50$ K of neighboring spins.

It is obvious that different configurations of spins and holes should also be energetically different. Besides their different magnetic ground state energies resulting from (7) they also differ by their Coulomb interaction between the holes and possibly by the Coulomb interaction with the local environment. In the following we take the electrostatic hole-hole repulsion by means of a screened Coulomb potential into account. $r_0 = 2.75$ Å is

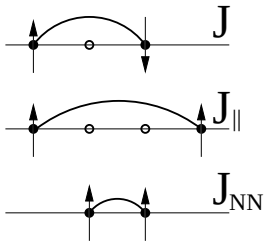

FIG. 4: Exchange parameters used for the exact diagonalization: $J = -134$ K, $J_{\parallel} = 14$ K, and $J_{NN} = 50$ K.

the distance between nearest neighbor sites on the ring. The accurate value of the dielectric constant ϵ_r is unknown. Several attempts have been undertaken to estimate the dielectric constant which yielded values for ϵ_r up to 30.[25,26,27] If the interaction with the local – modulated[15,28,29,30] – environment is neglected, it is necessary to assume a rather small dielectric constant $\epsilon_r \lessapprox 3$ in order to simulate the magnetization data.[17] Under such conditions only the energetically lowest-lying spin-hole configuration $\vec{c}$ contributes to the magnetization for $T \lessapprox 200$ K.

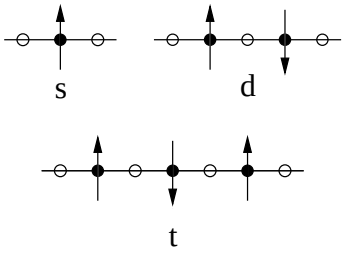

FIG. 5: Building blocks of the hole doped CuO_2 chains: open circles denote hole sites, arrows denote spin sites. The first block contains a single spin (s), the second a dimer (d), and the third block hosts three spins (t), which form a short chain.

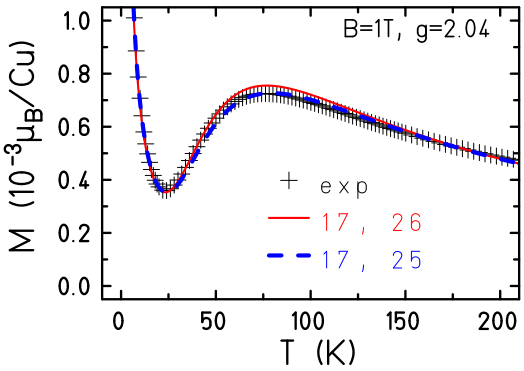

FIG. 6: (color online) Magnetization versus temperature for $B = 1$ T: Experimental data ($\vec{B} \parallel c$-axis) are given by crosses. The results of a complete numerical diagonalization are depicted by a solid curve for $N_s = 17$ and $N_h = 26$ as well as by a dashed curve for $N_s = 17$ and $N_h = 25$.

The assumption of a perfectly dimerized chain was studied in Ref. 17. In the following imperfect chains will be investigated. To this end Hamiltonian (6) is numerically completely diagonalized for the currently largest possible odd number of spins $N_s = 17$. The number of holes is chosen as $N_h = 25$ or $N_h = 26$ in order to yield about 60 % holes on the chain. Periodic boundary conditions are applied. It turns out that the ground state configurations are sequences of the building blocks shown in Fig. 5. In the case of $N_s = 17$ spins and $N_h = 26$ holes the resulting ground state sequence is $\vec{c}_{17,26} = sddddddddd$, i.e. it contains only dimers except for one single spin block. This ground state configuration is very similar to the one found using density functional theory calculations.[15,30] In the case of $N_s = 17$ spins and $N_h = 25$ holes the chain consists of dimers and one block of three spins, i.e. it has the sequence $\vec{c}_{17,25} = tddddddd$. In both cases other configurations of spins and holes are energetically well separated.

Having determined all energy eigenvalues and magnetic quantum numbers, the magnetization can be evaluated for the two ground state configurations. Figure 6 shows the resulting magnetization curves. The solid curve displays the magnetization curve for $N_s = 17$ and $N_h = 26$, i.e. configuration $\vec{c}_{17,26} = sddddddddd$, whereas the dashed curve shows the result for $N_s = 17$ and $N_h = 25$, i.e. $\vec{c}_{17,25} = tddddddd$. Both curves are rather close to the experimental data given by crosses.

IV. CONCLUSION

In this paper we present experimental as well as numerical evidence that the Curie-like contribution to the low-temperature magnetization of $Sr_{14}Cu_{24}O_{41}$ is a genuine property of the CuO_2 chain subsystem. The hole-doped chains are not regular sequences of weekly interacting spin dimers, but instead they host a small percentage of almost free spins and/or spin trimers. This finding is in accord with recent density functional calculations.[15,30] Nevertheless, our own simulational studies favor the occurrence of trimers instead of single spins which would be consitent with an intrinsic hole doping of the chains of less than 60 % as was inferred from recent NEXAFS experiments.[24]

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