

# Exact diagonalization studies of doped Heisenberg spin rings

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## Abstract

Motivated by magnetization studies of the “telephone number compound”  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  we investigate doped Heisenberg spin rings by means of complete numerical diagonalization of a Heisenberg Hamiltonian that depends parametrically on hole positions. A comparison with experimental magnetization data reveals rather accurate information about the screened electrostatic interaction between the charged holes on the ring which appears to be astonishingly strong.

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## 1. Introduction

Substances hosting spin and charge degrees of freedom promise a large variety of phenomena like magnetic and charge ordering, metallic conductivity and superconductivity [1,2]. The “telephone number compound”  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  contains two magnetic one-dimensional structures, chains and ladders. At low temperatures the ladder subsystem is magnetically inactive due to a large spin gap [3]. The stoichiometric formula  $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$  suggests 6 holes per formula unit. We will assume that for the undoped compound all holes are located in the chain subsystem, although this is experimentally under discussion since recent XAS measurements suggest that at room temperature some holes are located in the ladder subsystem [4] whereas it is necessary to assume that all holes are in the chain subsystem in order to explain neutron scattering data [5]. The coupling between the Copper spins depends on their exchange pathway across no, one, or two holes.

The magnetism of the chain subsystem (for various dopings) has been investigated for instance by means of classical spin dynamics [6,7] or by comparison with

spin-dimer models [8,9,10]. Usually the influence of the electrostatic hole-hole repulsion is neglected and a certain ground state configuration based on qualitative arguments is assumed. For the undoped compound the so-called dimer configuration is supposed to be the ground state configuration since this symmetric configuration should minimize the Coulomb repulsion. In addition, low-temperature susceptibility [10], neutron scattering data [5,11] as well as thermal expansion measurements [9] strongly support the existence of a low-lying dimer configuration.

The purpose of this article is as follows. We want to introduce a Heisenberg model which parametrically depends on hole positions. If the electrostatic hole-hole repulsion is included such a model allows to evaluate all energy eigenvalues and eigenstates (for small system size) and thus enables us to evaluate thermodynamic properties as function of temperature, magnetic field, and doping. Assuming certain exchange constants we can investigate the influence of the electrostatic hole-hole repulsion on ground state properties as well as on thermal averages like the magnetization which include contributions of low-lying spin-hole configurations.

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## 2. Model system

There are various ways to model a system of holes and spins, among them are the (multiband) Hubbard model and classical spin dynamics models. In this article the chain of holes and spins is modeled in the following way. 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 of a certain configuration is of Heisenberg type and depends parametrically on the actual configuration  $\mathbf{c}$ , i. e.

$$\hat{H}(\mathbf{c}) = - \sum_{u,v} J_{uv}(\mathbf{c}) \hat{\mathbf{s}}(u) \cdot \hat{\mathbf{s}}(v). \quad (1)$$

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). In (1)  $J_{uv}(\mathbf{c})$  are the respective exchange parameters. In this article three exchange parameters are considered:  $J = -67$  K is the antiferromagnetic coupling across a hole ( $J = -64$  K and  $J = -70$  K have been also considered),  $J_{\parallel} = 5.8$  K is the ferromagnetic coupling across two holes. The values of these exchange parameters are in accord with other theoretical and experimental investigations of this compound [10]. For the ferromagnetic coupling between spins not separated by a hole we are using  $J_{NN} = 8.7$  K, which is similar to the coupling used in Ref. [8]. A recent mean field analysis suggests that this exchange constant may be stronger [12].

The electrostatic interaction between holes is modeled by a potential energy

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

where  $r_0 = 2.75 \text{ \AA}$  is the distance between nearest neighbor sites on the ring and  $\epsilon_r$  is the dielectric constant. Several attempts have been undertaken to estimate the dielectric constant which yielded values for  $\epsilon_r$  up to 30 [13,14,15]. In related projects where the exchange interaction of chain systems in cuprates is derived from hopping matrix elements between different orbitals using a Madelung potential the dielectric constant is found to be 3.3 [16,17].

For small systems all hole configurations can be considered and the related spin Hamiltonians (1) can be diagonalized completely. For 8 spins and 12 holes this amounts to 6310 distinct hole configurations and tiny Hilbert spaces of dimension 256. For 12 spins and 18 holes the total number of hole configurations is already too big to be considered completely. Therefore, only the dimer configuration and low-lying excitations with their respective degeneracies are taken into account. It

will turn out that the high degeneracy of excited hole configurations plays an important role, since they substantially contribute to observables at low temperature although lying rather high in energy.

## 3. Numerical simulation and results

As an application we investigate spin rings where 60 % of the chain sites are occupied by holes. The ground state hole configuration for this compound is the so-called dimer configuration. In order to obtain this result it is necessary to consider the full Coulomb interaction. If one for instance tries to model the Coulomb interaction by a nearest neighbor repulsion, then the ground state is given by an antiferromagnetic chain and a cluster of the remaining holes, irrespective how big the nearest neighbor interaction is. Even the inclusion of next-nearest neighbors does not improve the situation, the Coulomb interaction is still proportional to the number of sites and may be overcome by the antiferromagnetic binding. We also find that the Coulomb interaction has to be sufficiently strong in order to yield the dimer configuration as the ground state. The following examples shows that this is indeed the case, the Coulomb interaction is only weakly screened.

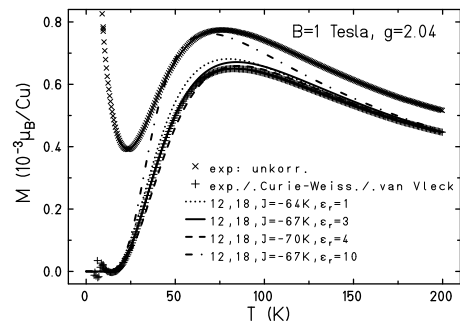


Fig. 1. Magnetization versus temperature for  $B = 1$  Tesla: Experimental data ( $\mathbf{B} \parallel c$ -axis) are given by x-symbols and crosses (corrected for Curie-Weiss and van-Vleck magnetism) [10]. For  $N_s = 12$  and  $N_h = 18$  magnetization curves are displayed for a range of exchange parameters  $J = -64, -67, -70$  K and several dielectric constants  $\epsilon_r$ .

Figure 1 shows the temperature dependence of the magnetization at  $B = 1$  Tesla for a system of 30 sites with  $N_s = 12$  spins and  $N_h = 18$  holes. The experimental data points for the infinite system ( $\mathbf{B} \parallel c$ -axis) are given by x-symbols and crosses (corrected for Curie-Weiss and van-Vleck magnetism) [10]. The latter can be very accurately approximated by an ansatz which describes the magnetic part as noninteracting antiferromagnetically coupled dimers [10]. Nevertheless, the additional ferromagnetic coupling  $J_{\parallel} = 5.8$  K as well

as other spin hole configurations also contribute to the magnetization and change the picture accordingly. If the Hamiltonian of the 12-spin system is diagonalized completely and Coulomb interaction is taken into account, one finds that the magnetization depends rather strongly on  $J$  and  $\epsilon_r$ . Figure 1 displays magnetization curves for a range of exchange parameters  $J = -64, -67, -70$  K and several dielectric constants  $\epsilon_r$ . For  $\epsilon_r = 1$  only the dimer configuration contributes. For  $\epsilon_r \gtrsim 3$  several hole configurations contribute with their respective magnetic spectra. For practical purposes only those configurations have been taken into account whose Coulomb energy differs by less than 500 K from the ground state Coulomb energy. It seems that a dielectric constant of the order of  $\epsilon_r \approx 3$  is best suited to describe the magnetization data. This result is in good agreement with a dielectric constant of 3.3 found in Refs. [16,17].

Figure 2 illustrates which configurations contribute for  $\epsilon_r = 3$  at lower energies. These configurations, although rather high in energy, nevertheless contribute with substantial weight, since they are highly degenerate. The degeneracy for configurations where one hole is moved is of order  $N_{\text{tot}}$ , the degeneracy for configurations where two holes are moved is of order  $N_s \cdot N_{\text{tot}}$ .

In order to estimate the influence of the finite size of the ring system we computed the magnetization for a larger ring with  $N_s = 16$  and  $N_h = 24$  which does not differ from the respective one with  $N_s = 12$  and  $N_h = 18$ , therefore it seems to be justified to assume that finite size effects do not play a role for the magnetization at this size.

#### 4. Outlook

In this article a Heisenberg model which depends parametrically on hole positions is introduced. It includes the electrostatic repulsion between holes and allows to evaluate thermodynamic properties as function of temperature, field, and doping. As a first application the influence of the electrostatic repulsion between holes on the temperature dependence of the magnetization is investigated. We find that the dielectric constant is approximately  $\epsilon_r \approx 3$  in order to reproduce the magnetization for 60 % holes on the ring.

Since the proposed model depends at least on four parameters ( $J, J_{\parallel}, J_{NN}, \epsilon_r$ ) the comparison to one magnetization curve, Fig. 1, leaves some freedom for the precise values. In future projects we are attempting to refine the model in order to obtain a consistent description of the meanwhile accumulated magnetization data. A direct measurement of the energy needed to excite hole movements, compare Fig. 2, would be very valuable since it would put additional restrictions on the range of the dielectric constant  $\epsilon_r$ . It is our hope

that a refined model will allow more insight into the interplay of charge order and magnetism.

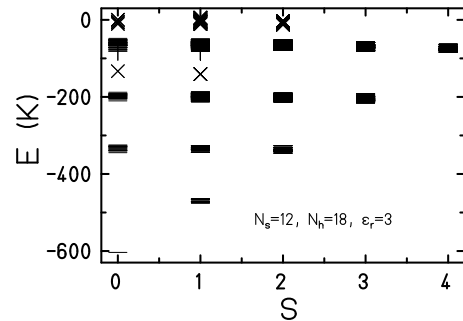


Fig. 2. Low-lying energy levels for  $J = -67$  K and  $\epsilon_r = 3$ : The dashes denote magnetic levels of the dimer-configuration, x-symbols show levels for configurations where one hole is moved, the crosses mark levels for two holes moved. The first triplet excitation is split into four levels with the following excitation energies (and degeneracies): 128 K (3), 131 K (6), 136 K (6), and 139 K (3).

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