Quantum Monte Carlo simulations for stacked spin-ladder systems containing low concentrations of nonmagnetic impurities: Application to the low-temperature broadening of NMR spectra in SrCu$_2$O$_3$

Robert Achleitner, Hans Gerd Evertz, Masatoshi Imada, Ralf Gamillscheg, and Peter Mohn

1Center for Computational Materials Science, Vienna University of Technology, Gussenhaußstraße 25/134, A-1040 Vienna, Austria
2Institute of Theoretical and Computational Physics, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria
3Department of Applied Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

(Rceived 3 October 2013; revised manuscript received 6 December 2013; published 23 December 2013)

We present a quantum Monte Carlo study for Heisenberg spin-$\frac{1}{2}$ two-leg ladder systems doped with nonmagnetic impurities. The simulations are applied to the doped spin-ladder compound Sr(Cu$_{1-x}$Zn$_x$)$_2$O$_3$, where a large broadening of the $^{65}$Cu NMR lines has been observed in experiment at low temperatures but far above the Néel temperature. We find that interladder couplings with a sizable coupling in the stacking direction are required to describe the line broadening, which cannot be explained by considering a single ladder only. Around a single impurity, spin correlations cause an exponentially decaying antiferromagnetic local magnetization in a magnetic field. We develop an effective model for the local magnetization of systems with many randomly distributed impurities, with few parameters which can be extracted out of quantum Monte Carlo calculations with a single impurity. The broadening arises from a drag effect, where the magnetization around an impurity works as an effective field for spins on the neighboring ladders, causing a nonexponentially decaying magnetization cloud around the impurity. Our results show that even for impurity concentrations as small as $x = 0.001$ and $x = 0.0025$, the broadening effect is large, in good quantitative agreement with experiment. We also develop a simple model for the effective interaction of two impurity spins.

DOI: 10.1103/PhysRevB.88.214422

PACS number(s): 05.30.–d, 75.10.Pq, 76.60.–k

I. INTRODUCTION

SrCu$_2$O$_3$ is a spin-$\frac{1}{2}$ Heisenberg spin-ladder compound that has been studied intensely both experimentally and theoretically. The crystallographically determined structure consists of planar Cu-O trellis lattices with intercalated Sr ions. These trellises contain the spin ladders which are almost perfectly decoupled due to frustration. The unpaired electron moment profiles around this impurity.5–13 However, fitting the exponentially decaying staggered effective local magnetic moment profile around this impurity.5–13

For BiCu$_2$PO$_6$, a differently structured material with large interladder coupling inside one layer, an exponentially decaying cloud model (\(\sim \exp\left[-(\xi_z/r_z + \xi_y/r_y)\right]\)) and a stacked ladder version (\(\sim \exp\left[-(\xi_x/r_x + \xi_z/r_z + \xi_y/r_y)\right]\)) are reported to show qualitative agreement with experiment for impurity concentrations around $x = 0.02$, but fail to explain the broadening at very small dilutions ($x \leq 0.005$).

In the present paper we perform quantum Monte Carlo (QMC) simulations for single and stacked spin ladders within the parameter range suggested in the report by Johnston et al. We develop an effective model for the magnetization of systems of stacked ladders with random impurities which needs only a few parameters measured by QMC and permits an efficient calculation of the NMR spectrum. We find that the coupling of adjacent stacked spin ladders strongly influences the NMR spectrum, and a value at the upper end of the range suggested in Ref. 16 is required to describe the experimentally found low-temperature NMR line broadening down to very small impurity concentrations, consistent with a study on the chain material Sr$_2$Cu$_4$O$_7$.

In Sec. II, we briefly summarize the results of NMR experiments on SrCu$_2$O$_3$ with nonmagnetic impurities. In Sec. III, we specify the Heisenberg model which we use to describe this material. Section IV contains results on single ladders, including an effective model for the interaction of two impurities, and Sec. V contains our results on stacked ladders. Section VI discusses the effects on NMR spectra, and Sec. VII contains our conclusions.

II. NMR EXPERIMENT

In their experimental investigation, Fujiwara et al. reported NMR studies on undoped and doped SrCu$_2$O$_3$. The results are shown in Fig. 1. For a doping of $x = 0.0025$ a massive broadening of the Cu NMR spectrum is observed upon lowering the temperature, while in the undoped specimen this effect is barely changes upon introduction of a few impurities.8,9,11,12

For BiCu$_2$PO$_6$, a differently structured material with large interladder coupling inside one layer, an exponentially decaying cloud model (\(\sim \exp\left[-(\xi_z/r_z)\right]\)) and a stacked ladder version (\(\sim \exp\left[-(\xi_x/r_x + \xi_z/r_z)\right]\)) are reported to show qualitative agreement with experiment for impurity concentrations around $x = 0.02$, but fail to explain the broadening at very small dilutions ($x \leq 0.005$).
almost absent. Similar experimental results have been obtained for SrCu2O3 by Ohsugi et al.,4 and also for other spin-ladder systems YBa2(Cu1−xZnx)3O6+y,18,19 YBa2(Cu1−xZnx)2O8,20 and Bi(Cu1−xZnx)2PO6.21

NMR measures the distribution of magnetizations in a system exposed to an external magnetic field. We note that quantitatively, the broadening of the NMR signal corresponds to relatively small magnetizations on the order of 0.001 ... 0.01 μB (see Sec. VI A). At first sight an increasing linewidth for decreasing temperature is counterintuitive. It suggests that the observed broadening is due to the temperature dependence of correlations along and between the spin ladders, and Refs 3 and 4 phenomenologically described the broadening with very large spin correlation lengths. However, the broadening takes place at temperatures far above the 3d ordering temperature of about 1 K in SrCu2O3 at the same impurity concentration,22,23 where long-range correlations would be expected.

In the following we will therefore investigate the distribution of magnetizations in a model for SrCu2O3 with impurities. We find that the spin correlation length is indeed barely affected by impurities in the temperature range of the NMR experiments, but the magnetization profiles around impurities is strongly influenced by the coupling of stacked ladders.

III. MODELING SrCu2O3

The unsaturated spins of the Cu2+ ions in the Cu-O planes in SrCu2O3 form spin ladders which are visualized in Fig. 2, where the spins along the ladder couple antiferromagnetically. Within the Cu-O planes the ladders form a trellis lattice, while normal to the Cu-O planes the ladders are stacked. The trellis lattice is responsible for an effective decoupling of the ladders due to frustration. We model SrCu2O3 like in Refs. 16 and 25 as a system of stacked ladders with spin-1/2 Heisenberg interactions,

\[
\hat{H} = k_B \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j - \mu_B g H \sum_i \vec{S}_i^z, \tag{1}
\]

with nearest-neighbor couplings \(J_{ij}\) and an external magnetic field \(H\), where the presence of an impurity still is useful even at large \(J_L\), where the presence of an impurity still breaks a singlet and leaves a free spin.

In a magnetic field, the antiferromagnetic correlations on the ladder cause a staggered local magnetic moment profile to
impurity, at two different temperatures. Upper curve (red/blue)
value) of a single ladder for JR/JL
of the system from responding to a small external field. An impurity
(left) an impurity. Spins form singlets with a spin gap which prevents
at large distances. By plotting the difference of the total magnetization
causes a temperature-dependent shift of the NMR peak without
changing its width, in quantitative agreement with the NMR
results in Fig. 1. The second effect is the creation of a local cloud of magnetic moments around the impurity [Figs. 4 and 5
insets]) which causes the total magnetization to grow again at
sufficiently low temperatures, corresponding to a shift back of the
NMR signal in Fig. 1 at temperatures of 40 K (0.02J_L) and
below. By plotting the difference of the total magnetization
between the undoped and the doped systems we find that the
total magnetization caused by the impurity corresponds to one
spin-1/2 moment5,9,35 at low temperature, in agreement with
experiment,22 and that it closely follows the analytical solution of a free spin in a magnetic field (dashed red line in Fig. 5)
M_{total} \approx g \langle S_{\text{free}}^z \rangle, \quad \text{(3)}
Correspondingly, the exponentially decaying clouds around
impurities on independent ladders shown in Fig. 4 produce
only a very small broadening of the NMR signal (see Sec. VI A).

B. Temperature and magnetic field dependence

We analyze the temperature dependence of spin
magnetizations9 in systems with a single impurity and without
impurities. We observe two mechanisms which have a direct impact on the NMR spectrum. In Fig. 5, both effects are
demonstrated for the total magnetization M_{total} = g \sum_{ij} \langle S_{ij}^z \rangle
of the doped and undoped systems. The undoped system (blue
up-triangles) exhibits a collective excitation of all the spins as
a response to the external field at temperatures above about
0.04J_L \approx 80 K (blue line in Fig. 5). This collective excitation
causes a temperature-dependent shift of the NMR peak without
changing its width, in quantitative agreement with the NMR
results in Sec. VI A.

FIG. 3. Rung picture of a spin ladder without (left) and with
(right) an impurity. Spins form singlets with a spin gap which prevents
the system from responding to a small external field. An impurity
breaks one singlet and leaves a free spin-1/2 which responds to small
external fields and couples to surrounding spins.

FIG. 4. (Color online) Local spin expectation value (absolute
value) of a single ladder for J_R/J_L = 0.5, H = 0.01J_L with a single
impurity, at two different temperatures. Upper curve (red/blue) T =
0.02J_L \approx 40 K, lower curve (black/green) T = 0.05J_L \approx 100 K. The
different colors refer to magnetic moments located on the undoped
ladder leg (red and black circles) and on the doped ladder leg (blue
and green triangles), respectively. For the two temperatures considered,
the correlation length remains almost constant (lower inset). The
upper inset shows the spin expectation values at T/J_L = 0.02 for
different values J_R/J_L. The correlation length \xi_i(J_R/J_L) becomes
5.9, 7.45, and 9.75 for J_R/J_L = 0.6, 0.5, and 0.4, respectively.

FIG. 5. (Color online) (a) Total magnetization versus T/J_L for a
system in a magnetic field, without (blue up-triangles) and with
(black circles) an impurity. System parameters: 200 \times 2 ladder, J_K/J_L = 0.5, H = 0.005J_K (\approx 14.3 T). To observe the effect caused by the impurity
only, the difference between the undoped and doped systems is also
shown (green down-triangles). The red dashed line is the analytical
solution for a free spin in a magnetic field. Blue and black lines are
guides to the eye. In the insets, the staggered distributions of local
magnetic moments around the impurity at (b) T = 0.005J_K \approx 9.6 K
and (c) T = 0.09J_K \approx 173 K are plotted. (For each inset: ladder leg
with impurity on the right, leg opposite impurity on the left.)
independent of the size of couplings, in the range studied. Indeed, we find that the individual local magnetic moments also show a tanh behavior, modulated by the exponential decay of Eq. (2).

\[ \langle S_z^i(T) \rangle = A(\xi_x)(-1)^{i+j} e^{-\frac{\mu_B H}{k_B T}} \]  

with a proportionality factor \( A(\xi_x) \) which depends on the correlation length.

In the temperature range \( T < 0.05 J_L \approx 100 \text{ K} \) where most of the broadening takes place in experiment, \( \xi_x \) is independent of temperature (Fig. 4, lower inset). In this range, \( A(\xi_x) \) can be obtained from a single QMC simulation by measuring \( \langle S_z^0 \rangle \) at a reference temperature \( T_{\text{ref}} \) and a magnetic field \( H_{\text{ref}} \):  

\[ A(\xi_x) = \frac{\langle S_z^0(T_{\text{ref}} / J_L) \rangle}{\frac{1}{2} \tanh \left( \frac{\mu_B H_{\text{ref}}}{k_B T_{\text{ref}}} \right)}. \]  

(5)

Figure 6 shows a comparison between the analytical model Eq. (4) and QMC results for three different lattice sites on the ladder leg opposite to the impurity (\( j = 0 \)). \( A(\xi_x) \) was extracted from a single QMC simulation at \( T_{\text{ref}} = 0.005 J_L \approx 10 \text{ K} \). In the broadening temperature regime (\( \xi_x \sim \text{const.} \)) and even beyond, the analytical model (dashed lines) matches the QMC data very well.

On the ladder leg containing the impurity (\( j = 1 \)), the magnitude of local spin expectation values is somewhat smaller than specified in Eq. (4) (see Fig. 4), consistent with the total homogeneous magnetization of the ladder to be that of a single free spin without a factor \( A \). [Eq. (3) and Fig. 5].

In Fig. 7 we compare Eqs. (3) and (4) to QMC results as a function of magnetic field \( H \) at fixed temperature. The match to local spin expectation values (inset) is very good. QMC results for the total magnetization (blue triangles) match the free spin solution perfectly for fields \( H / J_L \leq 0.07 \). Above \( H / J_L = 0.07 \) (corresponding to an applied magnetic field of about 200 T) the magnetization starts to rise significantly, which indicates that the applied magnetic field is large enough to break up the singlets. In simulations of the NMR response (discussed below), the magnetic fields never exceeds \( H / J_L = 0.01 \), so that Eq. (4) remains valid.

**C. Two impurities: Effective interaction**

The present work is primarily concerned with small impurity concentrations \( x \leq 0.0025 \). We therefore focus our investigation of impurity interaction on pairs of impurities, since for combinatoric reasons already three close impurities occur with very small probability.

We find the magnetic moment profile of two impurities to be close to a simple superposition of single-impurity profiles, but with a modified overall amplitude \( \langle S_z^0 \rangle_{\text{2imp}} \) instead of \( A(\xi_x) \). At small temperatures where Eq. (5) is valid, we find that the \( T \) and \( H \) dependence is again just a tanh, and with Eq. (4) the single-impurity profiles to be superposed are  

\[ \langle S_z^i(T) \rangle_{\text{2imp}} = \langle S_z^0(H_{\text{ref}} / T_{\text{ref}},d) \rangle_{\text{2imp}} \times (-1)^{i+j} e^{-\frac{\mu_B H}{k_B T_{\text{ref}}} \tanh \left( \frac{\mu_B H_{\text{ref}}}{k_B T_{\text{ref}}} \right)} \]  

where \( d \) is the distance between the impurities in the \( x \) direction.

\[ \langle S_z^i(H_{\text{ref}} / T_{\text{ref}},d) \rangle_{\text{2imp}} = \langle S_z^i(0 \text{ or } 0,0) \rangle_{\text{QMC}} / \left( 1 + e^{-\frac{\mu_B H_{\text{ref}}}{k_B T_{\text{ref}}} \tanh \left( \frac{\mu_B H_{\text{ref}}}{k_B T_{\text{ref}}} \right)} \right) \]  

for \( \langle S_z^i(0 \text{ or } 0,0) \rangle_{\text{QMC}} \) is measured by QMC calculation for 2 impurities. The normalization is such that the superposition will reproduce \( \langle S_z^i(0 \text{ or } 0,0) \rangle_{\text{QMC}} \). Figure 8 illustrates the quality of this description by plotting Eq. (6) together with QMC results for two different impurity distances such that the impurities reside on (i) the same and (ii) different sublattices of the underlying antiferromagnetic structure. Changing the impurity distance by one lattice site and thus changing the sublattice leads to very different magnetization profiles. This is a consequence of the antiferromagnetic order around the impurities, which can be described by two ferromagnetic sublattices that are shifted by one lattice site with respect to each other. If the impurities are an even number of lattice sites.

![Fig. 6.](image-url) FIG. 6. (Color online) Spin expectation values of the spins opposite of the impurity (\( i = 0 \)) and at a distance of 10 lattice sites (\( i = 10 \)) and 20 lattice sites (\( i = 20 \)) versus \( T / J_L \), all on the ladder leg opposite the impurity. The dashed lines correspond to a free spin in a magnetic field times a proportionality factor. System parameters: 200 x 2 ladder, \( J_R / J_L = 0.5 \), \( H / J_L = 0.005 \). The inset shows the factor \( A(\xi_x) \), measured at \( T = 0.02 J_L \).

![Fig. 7.](image-url) FIG. 7. (Color online) Total magnetization versus \( H / J_L \) for a system with a single impurity (blue up-triangles). System parameters: 300 x 2 ladder, \( J_R / J_L = 0.5 \), \( T_{\text{ref}} / J_L = 0.02083 \). The red dashed line corresponds to the solution of a free spin in a magnetic field. The inset compares Eq. (4) with the QMC results for \( \langle S_z^0 \rangle_{\text{QMC}} \) with \( A(\xi_x) \) measured at \( H_{\text{ref}} = 0.01 J_L \) and the same \( T_{\text{ref}} \).

![Diagram](image-url)
apart (same sublattice) their magnetization profiles enhance each other (Fig. 8, upper curve). If the impurity distance is an odd number (different sublattice), the profiles interfere destructively leading to the lower curve in Fig. 8. This latter case also resembles the formation of a domain wall.

The unpaired spins interact with each other via the staggered interaction, which decays exponentially with the spin correlation length. One may try to describe their interaction with an effective two-site Heisenberg model \(8,36–38\) for the two spins located on the same rungs as the impurities, with coupling \(J_{\text{eff}}\) and a magnetic field \(H\). We use the ansatz

\[
J_{\text{eff}} = C \left( J_R / J_L \right) (d^{-1} \exp(-d^{-1}/\xi_x)).
\]

Figure 9 compares the spin expectation values resulting from this model to QMC results for \(\langle S_i^z(d) \rangle_{2\text{imp}}\) as a function of distance \(d\) at different \(J_R/J_L\). We find excellent agreement down to very small distances for the case of odd distances, where the two spins form effective singlets. The fitted constants \(C(J_R / J_L)\) are 0.19, 0.245, and 0.29 for \(J_R/J_L = 0.4, 0.5, 0.6\), respectively. For the FM case the agreement is good at large distances, while for small \(d\), the two-spin model saturates whereas the magnetization on the full ladder continues to increase. Overall, the simple model Eqs. (6) and (8) describes the magnetization data in Figs. 8 and 9 very well.

V. STACKED LADDERS

A. Effective model, single-impurity case

QMC calculations of up to 8 stacked ladders have been performed using the parameters from Johnston et al. \(^{16}\) In these systems there is an interaction of spins on neighboring ladders via the exchange constant \(J_3\). Spins are now denoted as \(S_{i,j,k}\), where \(k\) is the distance in stacking direction from the ladder containing the impurity.

Figure 10 shows the spin expectation values on a six-ladder stack with increasing interaction strength \(J_3\) and a fixed ratio \(J_R/J_L = 0.5\). We use periodic boundary conditions in stacking direction; it is thus sufficient to plot only three ladders. For \(J_3/J_L \lesssim 0.001\) the stacked ladders behave almost like independent single ladders; the observed tiny magnetization variations in the neighboring ladder are within the numerical noise. With growing \(J_3/J_L\), an increasing influence on the neighboring ladders is seen. Ladder 1 which contains the impurity keeps a simple exponential behavior, with a slowly increasing correlation length, from \(\xi_x = 7.45\) at \(J_3 = 0\) to \(\xi_x = 9.75\) at \(J_3/J_L = 0.03\). However, the effective local magnetic moment distributions on the neighboring ladders do not follow a simple exponential decay law. Instead, the cusp which appears at \(S_0^z\) on the central ladder becomes progressively smeared out on neighboring ladders.

In Fig. 11 the spin expectation values \(|\langle S_{0,0,0}^z \rangle|\) are plotted for different \(J_3/J_L\) in stacking direction, from QMC calculations with 8 stacked ladders. With increasing \(J_3\) the deviation from a simple exponential decay increases. This deviation is illustrated in the inset of Fig. 11 where \(|\langle S_{0,0,0}^z \rangle|\) is compared

---

**FIG. 8.** (Color online) Magnetic moment profiles with two impurities on the same ladder leg. Full lines: description according to Eq. (6), symbols: QMC results. Upper curve: impurities are located at positions 123 and 137 on the same sublattice. Lower curve: impurities are located at positions 123 and 136 on different sublattices. QMC simulation for \(J_R/J_L = 0.5\), \(H_{\text{ext}} / J_L = 0.01\), \(T_{\text{ext}} / J_L = 0.02083\).

**FIG. 9.** (Color online) \(\langle S_i^z(d) \rangle_{2\text{imp}}\) Versus distance of two impurities on the same ladder leg. QMC results (black error bars) are compared to an effective two-site Heisenberg model (red triangles). The blue dotted line corresponds to a simple exponential superposition of two magnetic moment profiles calculated from a single impurity. \((T/J_L = 0.02083, H/J_L = 0.01, \text{system size } 200 \times 2)\)

**FIG. 10.** (Color online) Absolute values of the local spin expectation values induced by a single impurity \((\pm 0.083%)\) for different \(J_3/J_L\) with \(J_R/J_L = 0.5\). The system size is \(100 \times 2 \times 6\) spins with periodic boundary conditions in leg and stacking direction. \(T = 0.02083J_L \simeq 40\) K and \(H = 0.01J_L \simeq 30\) T. The impurity resides on the right leg \((j = 1)\) of ladder 1 \((k = 0)\). For the magnitude of spin expectation values see the inset, where the legs with \(j = 0\) of the same three stacked ladders are plotted at \(J_R/J_L = 0.019\) on a logarithmic scale, demonstrating the nonexponential behavior found on neighboring ladders.
with a cosh for $J_3/J_L = 0.02$. We find that for distant ladders the deviation from the hyperbolic cosine behavior increases. It thus becomes obvious that a simple exponential cloud model cannot be used to describe the complete profile caused by an impurity in a stacked ladder system.

Instead we developed a simple effective model, related to the one used in Refs. 17 and 39, for the actual distribution of impurity in a stacked ladder system. The ladder containing the impurity ($k = 0$) shows essentially the same profile as a single ladder and can therefore be described by Eq. (4) (with different normalization).

For the other ladders, let us first look at legs with $j = 0$ (opposite the impurity) We find that the spin expectations value $\langle \hat{S}_0,0,k \rangle$ on the next-neighboring ladder ($k = 1$) can be calculated by treating the spin on each site of the $k = 0$ ladder as a separate source of magnetization, with an exponentially decaying cloud in leg direction around it on the $k = 1$ ladder. Subsequently, the magnetizations on the $k = 2$ ladder are determined in the same way from those at $k = 1$, and so on for larger $k$. Thus, our ansatz is

$$\langle \hat{S}_l,0,k \rangle = e^{-\kappa_2} \sum_{l=0}^{L_x} (-1)^{l+1} \langle \hat{S}_l,1,0,k-1 \rangle e^{-\frac{\xi_2}{\kappa_2}}, \quad (9)$$

where $L_x$ is the length of the ladders and $\langle \hat{S}_l,0,0 \rangle$ can be computed like in Eq. (2). This approach treats the magnetization on ladder $k$ like a constant field for the spins on neighboring ladders, without back-action because of the small value of $J_3$. It describes the three-dimensional clouds of magnetizations around impurities very well, as we show below.

The strength of the coupling between the ladders is taken to be a ladder-independent factor $\exp(-1/\kappa_2)$ with an effective correlation length $\kappa_2$. Equation (9) is to be applied iteratively in $k$, up to the middle ladder in stacking direction, with the remaining ones determined by symmetry

$$\langle \hat{S}_{i,j,-k} \rangle = \langle \hat{S}_{i,j,k} \rangle. \quad (10)$$
and \( \langle S^z_0(d) \rangle_{2\text{imp}} \). Only the last of these depends on impurity positions.

VI. NMR

A. Magnetic moment profile and calculation of the NMR spectrum

To obtain the Cr\(^{2+} \) NMR spectrum influenced by a magnetic moment profile one has to consider the spin-ion hyperfine coupling in the resonance condition\(^{40} \)

\[
\frac{\nu_{RF}}{\gamma_N} = H_0(i,j,k) + A_{HF} \mu_B g^z(S^z_{i,j,k}(H_0(i,j,k)) \rangle.
\]

\( H_0(i,j,k) \) is the value of the external field \( H_0 \) matching the resonance condition at site \( i,j,k \). Here, \( \nu_{RF} \) is the frequency of the RF field, \( \gamma_N \) is the nuclear gyromagnetic ratio, \( A_{HF} \) is the hyperfine coupling (\(-12\ T/\mu_B\) for SrCu\(_2\)O\(_3\)), and \( \langle S^z_{i,j,k}(H_0(i,j,k)) \rangle \) is the expectation value for the \( z \) component of a spin at site \( i,j,k \) induced by an external field \( H_0 \).

The dependence on temperature and magnetic field of the NMR spectrum is then obtained by collecting the values of the RF field, \( \nu_{RF} \), the nuclear gyromagnetic ratio, \( \gamma_N \), the hyperfine coupling in the resonance condition, and the magnetic moment profile one has to consider the spin-ion hyperfine coupling in the resonance condition\(^{40} \). Here, \( \nu_{RF} \) is the frequency of the RF field, \( \gamma_N \) is the nuclear gyromagnetic ratio, \( A_{HF} \) is the hyperfine coupling (\(-12\ T/\mu_B\) for SrCu\(_2\)O\(_3\)), and \( \langle S^z_{i,j,k}(H_0(i,j,k)) \rangle \) is the expectation value for the \( z \) component of a spin at site \( i,j,k \) induced by an external field \( H_0 \).

The NMR spectrum is then obtained by collecting the values of \( H_0(i,j,k) \) from all lattice sites \( i,j,k \) into a histogram. To account for the natural linewidth we convolute the histograms with a Gaussian of 0.02 T half linewidth.

Figure 13 illustrates the resulting NMR spectrum arising from two impurities on a single ladder, with magnetic field strengths as used in the experiment by Fujiwara et al. (Fig. 1). Note that the broadening seen in experiment corresponds to very small values of \( \langle S^z_{i,j,k} \rangle \), up to about 0.01. Larger spin expectation values, which occur on only few sites close to impurities, contribute only little to the histogram, in its

\[
\nu_{RF} = H_0(i,j,k) + A_{HF} \mu_B g^z(S^z_{i,j,k}(H_0(i,j,k)) \rangle.
\]

The NMR spectrum is then obtained by collecting the values of \( H_0(i,j,k) \) from all lattice sites \( i,j,k \) into a histogram. To account for the natural linewidth we convolute the histograms with a Gaussian of 0.02 T half linewidth.

Figure 13 illustrates the resulting NMR spectrum arising from two impurities on a single ladder, with magnetic field strengths as used in the experiment by Fujiwara et al. (Fig. 1). Note that the broadening seen in experiment corresponds to very small values of \( \langle S^z_{i,j,k} \rangle \), up to about 0.01. Larger spin expectation values, which occur on only few sites close to impurities, contribute only little to the histogram, in its

FIG. 13. (Color online) Relation between local magnetic moment profiles and the corresponding NMR spectrum. Left panel: Spin expectation values (left and right leg) of a ladder containing two impurities on the left leg. Right panel: The corresponding NMR spectrum reflects the histogram of spin expectation values. The experimentally observed broadening corresponds to small values of \( \langle S^z_{i,j,k} \rangle \) up to about 0.01.

![Picture of NMR spectra](image-url)

FIG. 14. (Color online) NMR spectra with three impurities at fixed positions. Left panel: Influence of the stochastic noise on the width. Impurities at positions \((7,1,2), (51,2,6), (73,1,5)\), with QMC simulations of 10k (red dashed line) and 500k (black solid line) sweeps. The inset shows the dependence of the FWHM on the number of steps. Right panel: Two NMR spectra of two systems only differing in impurity configuration (250k steps). Black solid line: same as left panel; blue dashed line: positions \((35,1,1), (52,1,4), (86,2,5)\). \( J_L/J_L = 0.5, J_S/J_L = 0.017, T/J_L = 0.02083, H/J_L = 0.003828 \).

Outliers. For the understanding of the NMR signal such large spin expectation values may therefore be treated more approximately, as we do in our model descriptions.

Figure 14 illustrates two effects which influence the calculated distribution. The left panel shows that stochastic noise of small spin expectation values in the QMC calculation produces an effective broadening. Therefore QMC calculations of high precision are necessary in order to get reliable NMR histograms. In the right panel of Fig. 14 we show the influence of different impurity positions on the linewidth and line shape, which demonstrates the necessity to average over a large number of impurity configurations.

If each impurity configuration needed to be simulated in a separate QMC simulation, the computational effort would be too large. Instead, we employ the effective analytical description given in Sec. V A, which allows us to calculate the NMR spectra for many impurity configurations on the basis of only a few parameters measured in QMC simulations. For each set of couplings \( J_R/J_L \) and \( J_S/J_L \) we calculated \( \xi_{Ti} \), \( \langle S^z_0 \rangle \), \( \langle S^z_{0,1} \rangle \) on a \( 200 \times 2 \times 6 \) system of coupled ladders with one impurity (\( 200 \times 2 \times 8 \) for \( J_S/J_L = 0.03 \)). We calculated \( \langle S^z_{0,1}\text{imp} \rangle \) and \( \langle S^z(d) \rangle_{2\text{imp}} \) for distances \( d = 1 \ldots 40 \) of two impurities on the same leg of a ladder at \( J_S/J_L = 0.01 \) on \( 200 \times 2 \times 4 \) coupled ladders. Since this calculation was very time consuming, we used the ratio between \( \langle S^z(d) \rangle_{2\text{imp}} \) and \( \langle S^z_{0,1}\text{imp} \rangle \) also at other values of \( J_S \). All QMC calculations for the NMR spectra were done at the reference values \( T_{ref} = 0.02083 J_L = 40 \text{ K} \) and \( H_{ref} = 0.01 J_L \approx 19 \text{ T} \).

B. Results

We used our effective model to generate NMR spectra for given impurity concentrations and for different couplings, based on the QMC simulations described above. 10\(^4\) random
Experimental values for the $^{65}\text{Cu}$ central peak from Fig. 1 are given.

The couplings are $J_R/J_L = 0.1920$ K, $J_3/J_L = 0.5$ with stacked ladder couplings $J_3/J_L = 0.01$ (red dot-dot-dashed line) and $J_3/J_L = 0.03$ (solid red line), compared to experiment (Fig. 1) (black dashed line). Sizeable broadening occurs only for the larger interladder coupling. At high temperature (340 K), all spectra are narrow. For reference, we show an undoped system (green dotted line, Gaussian line shape) and the doped system (blue dot-dashed line) at $J_3/J_L = 0.03$. The inset shows an expanded temperature range.

Vacancy configurations were generated on systems of up to $400 \times 2 \times 20$ sites for each set of couplings $J_R/J_L$ and $J_3/J_L$. The NMR spectrum was calculated for each vacancy configuration using our model, and the results were superimposed to obtain spectra comparable to realistic NMR signals.

In Fig. 15 we show results at $x = 0.25\%$ impurity doping for two different stacked ladder couplings $J_3/J_L = 0.01$ and $J_3/J_L = 0.03$. At high temperature (340 K), all spectra are similar to the pure Gaussian line shape; i.e., no broadening is visible, even at the larger ladder coupling $J_3/J_L = 0.03$. This is due to the suppression of magnetic moments by the

Figure 15 shows the FWHM (full width at half maximum) versus temperature for different interladder couplings $J_3$. The couplings are $J_R/J_L = 0.5$ and $J_3/J_L = 0.0$ (resulting in $\xi_x = 7.45$), $0.01$ ($\xi_x = 7.6$, $\kappa_z = 0.218$), $0.02$ ($\xi_x = 8.6$, $\kappa_z = 0.253$), $0.03$ ($\xi_x = 9.5$, $\kappa_z = 0.278$), as well as $J_R/J_L = 0.4$ with $J_3/J_L = 0.02$ ($\xi_x = 11.554$, $\kappa_z = 0.254$). We find that simulated spectra with a stacked ladder coupling slightly

At low temperature, $T = 40$ K, the NMR spectrum remains narrow for isolated ladders (not shown) since an exponential decay with the small correlation length of the undoped system (Fig. 4) does not contain enough sites with the relevant range of magnetizations. The spectrum remains narrow also for small interladder coupling $J_3/J_L = 0.01$.

In Fig. 16 we study the broadening at $x = 0.25\%$ in more detail. We show the FWHM (full width at half maximum) versus temperature for different interladder couplings $J_3$. The couplings are $J_R/J_L = 0.5$ and $J_3/J_L = 0.03$ and different temperatures compared to experiment (Ref. 3) ($^{65}$Cu left peak, $v_{RF} = 83.55$ MHz, filled circles).

The behavior changes drastically for the larger interladder coupling $J_3/J_L = 0.03$. Then several stacked ladders obtain magnetizations within the relevant range (Fig. 11), resulting in a broadened NMR spectrum in excellent agreement with the experimental spectrum.

In Fig. 17 we show the FWHM at $x = 0.0025$, $J_L = 1920$ K, $J_R/J_L = 0.5$, and $J_3/J_L = 0.03$ and different temperatures compared to experiment (Ref. 3) ($^{65}$Cu left peak, $v_{RF} = 125.1$ MHz, filled circles). The asymmetric experimental profiles below 40 K are caused by an overlap of a $^{65}$Cu transition with its main peak around 10.05 T. The experiment appears to have a larger natural linewidth than assumed in our simulations.
FIG. 19. (Color online) Simulated NMR spectra (dashed lines) at $x = 0.01$, with $J_L = 1920 \text{ K}$, $J_R/J_L = 0.5$, and $J_R/J_L = 0.03$ and different temperatures compared with experiment (Ref. 4) ($^{65}$Cu left peak, filled circles). The inset shows the same system with an impurity concentration of $x = 0.02$.

below 0.03$J_L$, at the upper end of the couplings specified in Ref. 16, match the experimental results well over the whole temperature range. The correlation lengths and $\kappa_z$ were extracted at $T = 0.02083J_L = 40 \text{ K}$ and assumed to be temperature independent. This assumption is valid for the low-dilution, low-temperature regime, while for higher temperature there is little broadening so that the influence of the correlation length becomes unimportant.

In Figs. 17, 18, and 19, we show a detailed comparison of our simulated NMR spectra with experimental results at impurity concentrations of 0.25%, 0.1% (which appears to have a larger natural linewidth), and 1%, using $J_R/J_L = 0.5$ and $J_R/J_L = 0.03$. We find that our model is in very good agreement with experiment in almost all cases.

VII. CONCLUSIONS

We studied impurity-containing systems of stacked spin ladders by means of QMC simulations. Temperature and field dependence of magnetizations are paramagnetic. We developed an effective spin model for the interaction of unpaired spins next to two impurities. In contrast to assumptions made in earlier investigations we observed that the staggered magnetization caused by an impurity does not follow a simple three-dimensional exponential behavior. The spin distributions on the ladders in stacking direction deviate progressively from such an exponential dependence. We provided an analytical description for the spin profiles in systems with multiple impurities and used it to simulate the NMR spectra of lightly doped SrCu$_2$O$_3$ with only a small number of parameters determined by QMC. The resulting NMR spectra allowed us to explain the drastic broadening of the $^{65}$Cu NMR line in SrCu$_2$O$_3$ found in experiments at intermediate temperatures to be a consequence of a sizable coupling between ladders in stacking direction, which causes a nonexponential cloud of small effective magnetic moments to occur around impurities.

ACKNOWLEDGMENTS

The authors acknowledge support from the Austrian Science Fund FWF within SFB ViCoM F4109-N13 P04 and P09 (H.G.E. and P.M.) and from the Science College W401-N13 (R.A.).


1920 K, $J_R/J_L = 0.5$, and $J_R/J_L = 0.03$. We find that our model is in very good agreement with experiment in almost all cases.
26 In specifying $H/J_L$ in the paper we leave out the unit tesla/kelvin, i.e., $H/J_L = 0.01$ should be read as $(\mu_B H)/(k_B J_L) = 0.01$.
33 Potential additional cyclic couplings (Ref. 41) are not accessible by QMC calculations because of a severe sign problem.