Saturation Field of Frustrated Chain Cuprates: Broad Regions of Predominant Interchain Coupling

S. Nishimoto,1 S.-L. Drechsler,1,* R. O. Kuzian,1 J. van den Brink,1 J. Richter,2 W. E. A. Lorenz,1 Y. Skourski,3 R. Klingeler,4 and B. Büchner1

1IFW Dresden, P.O. Box 270116, D-01171 Dresden, Germany
2Universität Magdeburg, Institut für Theoretische Physik, Germany
3Hochfeld-Magnetlabor Dresden, Helmholtz-Zentrum Dresden-Rossendorf, D-01314 Dresden, Germany
4Kirchhoff Institute for Physics, University of Heidelberg, D-69120 Heidelberg, Germany

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A thermodynamic method to extract the interchain coupling (IC) of spatially anisotropic 2D or 3D spin-1/2 systems from their empirical saturation field \(H_s(T = 0)\) is proposed. Using modern theoretical methods we study how \(H_s\) is affected by an antiferromagnetic (AFM) IC between frustrated chains described in the \(J_1-J_2\)-spin model with ferromagnetic 1st and AFM 2nd neighbor in-chain exchange. A complex 3D-phase diagram has been found. For Li\(_2\)CuO\(_2\) and Ca\(_2\)Y\(_2\)Cu\(_5\)O\(_{10}\), we show that \(H_s\) is solely determined by the IC and predict \(H_s \approx \) 61 T for the latter. With \(H_s \approx \) 55 T from magnetization data one reads out a weak IC for Li\(_2\)CuO\(_2\) close to that obtained from inelastic neutron scattering.

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Since real spin chain systems exhibit besides a significant in-chain coupling also an interchain coupling (IC), one may ask: in which cases is this relatively weak IC still important or even crucial? From the Mermin-Wagner theorem its decisive role for the suppression of fluctuations is well-known. The IC leads to long-range order at \(T = 0\) in 2D [1] and at \(T < T_N\) in 3D [2]. Often one is faced with a situation that the large in-chain couplings are known with reasonable precision, e.g., from inelastic neutron scattering (INS) or susceptibility data [3], but precise values for the tiny (nevertheless important) IC are lacking. Without magnetic frustration the IC can be determined quite accurately, e.g., from \(T_N\) analyzed by Quantum Monte Carlo studies [4]. But how to extract from experimental data a small IC for frustrated systems with weakly coupled chains where these methods do not work? Here we address such a 2D or 3D problem for the case of frustrated spin-1/2 chains with ferromagnetic (FM) 1st neighbor and antiferromagnetic (AFM) 2nd neighbor exchange described by the isotropic \(J_1-J_2\) model (IM). Nowadays it is the standard model for edge-shared chain cuprates (see, e.g., [5]). This 1D-IM attracted much interest [6–11] due to a rich phase diagram with multipolar (MP) phases derived from multimagnon bound states (MBS) in high magnetic fields [12–14]. Additional AFM degrees of freedom enhance the kinetic energy of magnons and AFM IC might disfavor multi-MBS. Hence, a precise knowledge of the magnitude of the IC is a necessary prerequisite to attack the multi-MBS problem, including a possible MBS Bose-Einstein condensation [15–18], and thus the IC is of general interest. Since Li\(_2\)CuO\(_2\) (see Fig. 1) is one of the best studied frustrated cuprates, it is well suited to compare theory and experiment. In particular, the main in-chain and IC \(J\)’s were extracted from INS data and a specific AFM IC was found crucial for preventing spiral order in the 3D ground state (GS) [3]. If the saturation field \(H_s\) would be known, the INS derived IC could be checked. But so far \(H_s\) has not been measured for Li\(_2\)CuO\(_2\). Here we report high-field magnetization data to fill this gap. Our paper is organized as follows. We recall the 1D case and provide details of the density-matrix renormalization group (DMRG) technique involved. Then we report results for coupled chains, including a complex phase diagram and a comparison with our experimental \(H_s\)-data for Li\(_2\)CuO\(_2\) fully explained in terms of a predominant IC.

We apply the DMRG method [19] with periodic boundary conditions (PBC) in all directions. Seemingly, this method is less favorable for \(D > 1\); however, on modern workstations using highly efficient DMRG codes, spin systems with up to about \(\sqrt{10} \times \sqrt{10} \times 50\) sites, i.e., 10 coupled chains of length \(L \approx 50\), can be studied. Thus, by taking a proper arrangement of the chains, 3D-lattices can be simulated cf. the inset in Fig. 2. Let us describe how the block states are constructed in an \(n \times L\) cluster, where \(n\)

![Image](https://example.com/image.png)

**FIG. 1** (color). Left: Crystal structure of Li\(_2\)CuO\(_2\) with two CuO\(_2\) chains per unit cell along the \(b\) axis. Right: View along the \(c\) axis on the \(ab\) plane. The main in- and interchain couplings \(J_{1,2}\) and \(J_{IC}\), \(J'_IC\): arcs and dashed lines, respectively. The normalized ICs read \(\beta_1 = J'_IC/J_1\) and \(\beta_2 = J'_IC/J_2\).
denotes the number of chains and \( L \) is the chain length. If we regard \( n \) sites in the \( ac \)-plane as a “unit cell”, the system can be treated as an effective 1D chain with \( L \) sites (step 1). This enables us to use an appropriate 1D array for the construction of the PBC (see Fig. 1 of Ref. [20]). In step 2 the sites within each “unit cell” are arranged into numeric order (see the inset of Fig. 2). Thus, the distance between most separated interacting sites can be held at 11 and 23 in the 4- and 8-chain systems, respectively. Since the interactions run spatially throughout the system, the wave function converges very slowly with DMRG sweep but without getting trapped in a “false” GS. We kept \( m = 1600–4000 \) density-matrix eigenstates. About 30 sweeps are necessary to obtain the GS energy within a convergence of \( 10^{-7} |J_I| \) for each \( m \) value. All calculated quantities were extrapolated to \( m \to \infty \) and the maximum error in the GS energy is estimated as \( \Delta E/|J_I| \sim 10^{-4} \), while the discarded weight in the renormalization is less than \( 1 \times 10^{-6} \). For high-spin states \([S_{tot} \approx (nL - 10)/2]\) the GS energy is obtained with an accuracy of \( \Delta E/|J_I| < 10^{-12} \) by carrying out several thousands sweeps even with \( m = 100–800 \). Then, we obtain the reduced saturation field \( h_s = g \mu_B H_s/|J_I| \) with high accuracy (e.g. 12 digits as compared to exact solutions available in some cases). The assignment in the 1D phase-diagrams [11–14,21] and in 3D (see Fig. 3) stems from the magnetization curves slightly below \( h_s \) (see Figs. 2 and 6). The signature of each phase is the height of the magnetization steps \( \Delta S^z = 1, 2, 3, \ldots \) for di- (1-magnon), quadru- (2-MBS), octu- (3-MBS), and hexadecapolar (4-MBS), etc., phases, respectively. For the 2-MBS case the DMRG results are confirmed by the exact hard-core boson approach (HCBA) [9].

We start with the 1D problem. With \( \alpha = J_2/|J_I| \) the critical point is given by a level crossing of a singlet and the

\[ \frac{g \mu_B H_s}{N_{IC} (J_{IC} + J'_{IC})} \quad \text{for} \quad J_{IC} \geq J'_{IC}, \]

where \( N_{IC} \) is the number of nearest interchain neighbors (8 for \( \text{Li}_2\text{CuO}_2 \)). At \( \alpha > 1 \) there is no C-phase. For simplicity [22] we take \( J'_{IC} = 0 \). Then, \( J_{IC}^{cr+} (\alpha)/J_I = (1-4\alpha)/9 \), if \( \alpha < 0.57 \) which is obeyed for \( \text{Li}_2\text{CuO}_2 \). The INC2-C transition in the interval \( 0.57 \leq \alpha < 1 \) is of 1st order [21]. For \( \text{Li}_2\text{CuO}_2 \) [3], we have \( J_{IC} = 9 \text{ K} > J_{IC}^{cr+} (0.332) = 0.0364 |J_I| = 8.2 \text{ K} \) [23]. Since in the
Quantum fluctuations \( \langle S \cdot S \rangle_{J_s} - \frac{1}{4} \) (b) in-chain and IC spin-spin correlation functions vs applied field, given by the filled symbols ■ and ●, respectively, from DMRG.

C phase \( H_s \) depends solely on \( J_{IC} \), the IC can be read off from experiment: \( H_s = 55.4 \) T yielding \( J_{IC} = 9.25 \) K very close to \( 9.04 \) K from zero-field INS data [3]. In the INC1 phase \( J_{IC} \) dominates \( H_s \). There above \( J_{IC}^{cr,+} = 0.0219\beta \) only INC 1-magnon low-energy excitations exist. Below \( J_{IC}^{cr,-} \) 3-MBS are recovered as low-energy excitations. The transition from the 3-MBS to the INC1-phase is 1st order. The general case is shown in Fig. 4(a). Well below \( J_{IC}^{cr,+} \), \( H_s \) significantly depends on \( \alpha \). The yellow stripe highlights the region of predominant IC addressed in our title. Here this dependence is weak. Subtracting their classical value, the spin-spin correlation functions show that the in-chain fluctuations vanish much faster than the IC ones for \( H \to H_s \) [see Fig. 4(b)] in accord with the surprising result of Eq. (1). In the FM region \( \alpha < 1/4 \) the in-chain contribution vanishes by definition. Hence, the external field has to overcome the AFM IC, only, and Eq. (1) is valid. This is the case of \( \text{Ca}_2\text{Y}_2\text{Cu}_6\text{O}_{10} \) [24] with an IC geometry like in \( \text{Li}_2\text{CuO}_2 \) and a 2D chain arrangement, i.e. \( N_{IC} = 4 \). Its reduced \( N_{IC} \) is overcompensated by a larger IC \( J_{IC} + J_{IC}^{cr} = 26 \) K [25]. Then we predict \( H_s = 61 \) T refining a value of 70 T overestimated from low-field data [24].

Pulsed-field magnetization studies have been performed at the Dresden High Magnetic Field Laboratory in fields up to 60 T. The results taken at \( T = 1.45 \) K for \( H \parallel b \) axis on a \( \text{Li}_2\text{CuO}_2 \) single crystal from the same batch as in the INS-study [3] are shown in Fig. 6. The data imply a quasi-linear increase of the magnetization \( M(H) \) between 10 and 30 T, i.e. \( \delta M/\delta H = M' = \text{const} \). Above about 50 T \( M' \) increases notably and pronounced peaks develop at \( 55.4 \pm 0.25 \) T and \( 55.1 \pm 0.25 \) T for two pieces of our single crystal. The sharp drop of \( M' \) towards 0 at higher fields justifies to attribute the peaks with \( H_s \). The saturation moment amounts to \( M_s = 0.99 \pm 0.06 \mu_B/\text{f.u.} \) using \( g_b = 1.98 \pm 0.12 \) in reasonable agreement with \( g_b = 2.047 \) from low-field ESR-data at 300 K [26]. In Fig. 6 we compare \( M/M_s \) from the DMRG with the experimental data for \( \text{Li}_2\text{CuO}_2 \). The DMRG description in this plot is

![Figure 4](image-url)

**Figure 4** (color). (a) Approximate in-chain contribution to the saturation field measured by its deviation from Eq. (1) for different \( \alpha \) values vs \( \beta_2 \) in units of its critical value \( \beta_2^{cr} \) (see Figs. 3, S1 of Ref. [21], and text). Yellow stripe: region of predominant IC addressed in our title. Here this dependence is weak. (b) in-chain and IC spin-spin correlation functions vs applied field, given by the filled symbols ■ and ●, respectively, from DMRG.

**Figure 5** (color). The saturation field \( h_s \) at \( \alpha = 1/3 \) vs \( \beta_2 \) from DMRG method. Notice the two critical IC values \( \beta_2^{cr} = 0.0109 \) and \( \beta_2^{cr} = 0.0374 \) denoted by dashed vertical lines. For INC1- and C- phases, see text. Inset: the weak nonlinearity of \( h_s \) vs \( \beta_2 \).

**Table I.** Saturation field \( h_s \) at \( \alpha = 0.332 \), \( \beta_2 = 3/76 \), and \( \beta_1 = 0 \). \( J_{IC} \) has been multiplied by 4 for 2-chain systems.

<table>
<thead>
<tr>
<th>( L )</th>
<th>single chain</th>
<th>2-chain system</th>
<th>8-chain system</th>
</tr>
</thead>
<tbody>
<tr>
<td>16</td>
<td>0.061 048 005 894 2</td>
<td>0.315 789 473 684</td>
<td>0.315 789 473 684</td>
</tr>
<tr>
<td>48</td>
<td>0.061 691 042 337 8</td>
<td>0.315 789 473 684</td>
<td>0.315 789 473 684</td>
</tr>
<tr>
<td>96</td>
<td>0.061 691 048 727 0</td>
<td>0.315 789 473 684</td>
<td>0.315 789 473 684</td>
</tr>
<tr>
<td>144</td>
<td>0.061 691 048 724 7</td>
<td>0.315 789 473 684</td>
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Hs accessible quantities, the Curie-Weiss temperature and above. Notably, linear relations of two experimentally

Li$_2$CuO$_2$

M

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Hs external fields and, particularly, the strength of H, quasi-1D systems, such as the applicability of the SWT. To extract confirm the validity of the adopted spin Hamiltonian and field data close to a value from previous INS study we obtained from several independent theoretical and experimental studies discard any 1D scenario for Li$_2$CuO$_2$, even more, H$_s$ itself is (within the isotropic model) independent of the in-chain couplings J$_1$ and J$_2$. Thus, our results show exactly that for a rather wide interval 0 < \alpha < 1 and collinear magnetic order at \( H = 0 \), H$_s$ depends only on the IC, irrespective of its strength. A complete study of the entire phase diagram including the INC2 phase will be given elsewhere. The MP-phases from 1D studies are very sensitive to the presence of IC in the 2D or 3D systems. In particular, they can be eliminated by a weak AFM IC. Instead new incommensurate phases may occur. A study of H$_s$ in other systems within the approach proposed here is in progress.

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*Corresponding author.
s.l.drechsler@ifw-dresden.de

[22] Its account is possible but electronic structure calculations suggest \( J_{IC}/J_1 \approx 0.1 \), only.
[23] From the error bars of \( J_1 \) and \( \alpha \): ±5 K and ±0.005, respectively, we have \( 7.18 \, \text{K} \leq J_{IC}^{\pm} = 8.54 \, \text{K} \), and \( J_{IC}^{\pm} \) is well below the \( J_{IC} \) values obtained from the INS or \( H_s \) data.