On spin-1/2 multipolar phases in frustrated quasi-one-dimensional systems with ferromagnetic nearest neighbor inchain exchange

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Antiferromagnetically (AFM) coupled frustrated spin-1/2 chains in magnetic fields are studied by the DMRG technique and spin wave theory. The exchange along the chains is described within the J_1-J_2 Heisenberg model with ferromagnetic (FM) first neighbor and AFM second neighbor couplings. The multipolar (MP) ground states especially for those related to 3-, 4- or higher magnon bound states (MBS) are destroyed by a weak AFM interchain coupling (IC). Details are of less importance. Based on band structure and empirical estimates for the IC we suggest that quantum spin nematics might be found for $LiVCuO_4$ whereas for $Li(Na)Cu_2O_2$ it is unlikely due to a relatively strong IC. Similarly even for a much smaller IC from 3- or 4-MBS derived MP phases are for Li_2ZrCuO_4 unlikely, too. Here a saturation field of ≈ 13 T is predicted. It much exceeds the 1D value of 4.7 T.

I. INTRODUCTION

Quasi-one-dimensional (Q-1D) spin chain compounds exhibit in addition to significant intrachain coupling always also an interchain coupling (IC). To understand their physics, especially with respect to that suggested by appropriate 1D-models for a single chain, the question arises in which cases even a relatively weak IC is still important or even crucial? The transition from 1D to 2D or 3D is often non-trivial. Features found in 1D might not occur there. In particular, from quantum mechanics it is well-known that bound or localized states are strongly pronounced in 1D and more rare in 2D or 3D. Here we address such a problem, namely, the fate of multipolar states related to multimagnon bound states (MBS) especially at high external magnetic fields. We consider frustrated edge-shared cuprates formed by spin-1/2 CuO_2 chains with ferromagnetic (FM) nearest-neighbor (NN) and antiferromagnetic (AFM) next-nearest-neighbor (NNN) exchange described approximately by the isotropic J_1-J_2-Heisenberg model[2]. Recently, this 1D-model and related compounds revealed considerable interest[3–16] due to a rich and exotic phase diagram with multipolar phases (MP)[17–19]. Nowadays it is the most popular model for edge-shared chain cuprates (see e.g.[6]). Additional AFM couplings typically provided by the IC enhance the kinetic energy of magnons. Hence, such an AFM IC might disfavor the formation of low-lying MBS. On the contrary, FM IC might stabilize such MP or create even new MP states. A realistic examination of real quasi-1D systems to evaluate the chances to detect remnants of the rich 1D physics, to find novel states or unusual properties, and to compare with experiments is of broad interest[11–13,20–22].

II. COMPOUNDS AND TYPES OF INTERCHAIN INTERACTION

Among edge-shared cuprates $LiCu_2O_2$, the isomorphic $NaCu_2O_2$, and $LiVCuO_4$ (see Fig. 1; for comparison the reference system Li_2CuO_2 is shown, too) have been proposed to be candidates for quantum-spin nematics, i.e. quadrupolar phases derived from 2-MBS. Other systems like Li_2ZrCuO_4[16] and $Rb_2Cu_2Mo_3O_{12}$ located closer to the FM-spiral critical point might be regarded as candidates for the triatic or quartic MP related to higher, i.e. to 3- and 4-MBS. Besides the strength of the IC also the influence of various topologies of the IC resulting from different 3D arrangements of individual chains is of interest. The main types of IC are depicted in Fig. 1. The simplest case is given by unshifted neighboring chains and a predominant perpendicular IC. In this situation spirals on neigboring chains are only weakly affected by an AFM IC[11] (classically their pitch angle, i.e. the incommensurate (INC) magnetic structure along the chains in the dipolar phase at ambient external fields remains even unaffected). Hereafter, we call this IC the 'unfrustrated IC'. Even with four CuO_2-chains per unit cell, only an effective 2D chain arrangement is realized approximately for $Li(Na)Cu_2O_2$, where $J_{ic} \sim$(0.5 to 1)$J_2 \sim$ 40 to 100 K[25–27]. A square lattice of unshifted chains, i.e. a simple 3D case considered in Ref. 22 is not realized in cuprates due to the anisotropic electronic (orbital) structure of the basic CuO_4-plaquettes. Nevertheless, we will refer to that academic case (but most frequently considered in the theoretical literature on quasi-1D systems) to compare the theoretical methods and approximations applied there with our approach (see Fig. 2). (Another 3D case is given by a triangular lattice of chains. It occurs in hexagonal structures and it is also of less interest for frustrated edge-shared cuprates.) A bit more complex 3D case but with shifted neighboring chains is realized for Li_2CuO_2(see Fig. 1). The strength of the resulting frustrating AFM IC has been found crucial for preventing a spiral order in the 3D ground state[12]. For that well-studied reference compound $H_s \approx 55.5$ T has been measured recently[13]. From this data, from inelastic neutron scattering (INS) studies, and from recent band-structure calculations the IC can be determined with high accuracy[12]. As a result one arrives at 1 meV as the relevant IC energy scale. This holds also for Li_2ZrCuO_4[9] with buckled chains which show a reduced IC.

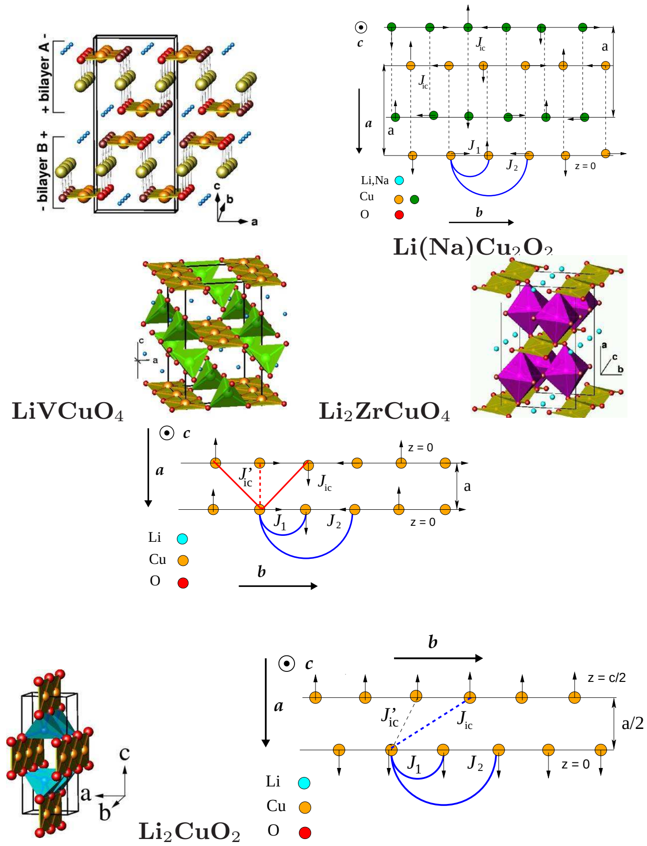

FIG. 1: (Color) Crystal structure and exchange pattern of chain cuprates discussed in the text. The main in- and interchain exchange paths, $J_{1,2}$, $J_{\rm ic}$, and $J'_{\rm ic}$ are marked by arcs and dashed lines, respectively. Upper: On the right a projection of half of the unit cell with two of four chains is shown. The depicted planes with green and yellow Cu sites are considered as uncoupled in the present simplified analysis. Only perpendicular (unfrustrated) IC in each plane is taken into account. Middle: The main interaction in the basal plane of unshifted chains. The IC for the first and second diagonal (not shown) are frustrating in the sense of spirals as discussed in the text. Lower: Projection of two NN shifted chains in different (ab)-planes stacked along the c-axis. Here the frustrated interaction between these planes provides the leading IC. The first diagonal IC $J'_{\rm ic}$ is much weaker than the second one $J_{\rm ic}$ we take into account here, only (see also Ref. 12).

III. THEORETICAL METHODS

We used the density-matrix renormalization group (DMRG) method[23] with imposing periodic boundary conditions (PBC) for all directions. In general, it is

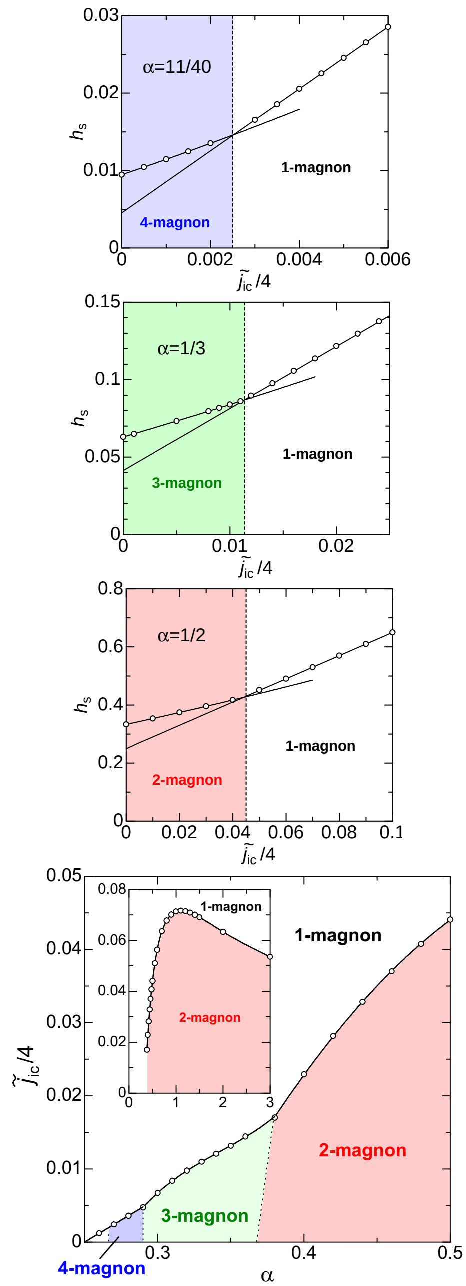

FIG. 2: (Color) The critical interchain coupling (IC) for the 3D case of a square lattice arrangment od chains with unfrustrated perpendicular renormalized IC $\tilde{j}_{\rm ic} = N_{\rm ic} J_{{\rm ic}\,\perp} / \mid J_1 \mid$ vs. inchain frustration rate $\alpha = J_2/J_1$, where $N_{\rm ic} = 4$ is the number of nearest interchain neighbors in 3D. The correpondiing renormalized 2D case is rather similar apart from slightly larger quantum fluctuation more pronounced in case of less neigbors $N_{\rm ic} = 2$ (see also the remark in Fig. 3)

known that this method is much less appropriate for $D > 1$. However, spin systems with up to about $\sqrt{n} \times \sqrt{n} \times L = \sqrt{10} \times \sqrt{10} \times 50$ sites can be studied by taking a proper construction of the lattice block (See Ref. 13). We kept $m \approx 800 - 4000$ density-matrix eigenstates in the renormalization procedure. In fact, about $100 - 300$ sweeps are necessary to obtain the GS energy within a convergence of $10^{-7} J_1$ for each m value. All calculated quantities were extrapolated to $m \to \infty$ and the maximum error in the GS energy is estimated as $\Delta E/J_1 \sim 10^{-4}$, while the discarded weight is less than 1×10^{-6}. Under the PBC, a uniform distribution of $\langle S_i^z \rangle$ may give an indication to examine the accuracy of DMRG calculations for spin systems. Typically, $\langle S^z \rangle - S^z_{\rm tot}/(nL)$ is less than 1×10^{-3} in our calculations. Note that for high-spin states $[S^z_{\rm tot} \gtrsim (nL - 10)/2)]$ the GS energy can be obtained with an accuracy of $\Delta E/J_1 < 10^{-12}$ by carrying out several thousands sweeps even with $m \approx 100 - 800$. As a result, we obtain the reduced saturation field $h_s \equiv g\mu_{\rm B} H_{\rm s}/|J_1|$ with high accuracy. In addition to DMRG we have also applied the linear spin wave theory and the hard core boson approach[10]. The latter provides exact results for the 2-MBS phases. Some qualitative results for the reported magnetization curves have been additionally checked applying the complete diagonalization of finite chain systems. All methods do provide in the regions of their validity practically coinciding results. Some examples are given in Ref. 13.

IV. RESULTS

A. Unfrustrated interchain couling and unshifted chains in 2D and 3D

We start with the simplest IC model of a 2D and the simplest 3D ordering of chains within a square-lattice arrangement of parallel chains with a single AFM perpendicular IC (called also "unfrustrating" as explained above), only. This model provides a reasonable 2D approximation for $Li(Na)Cu_2O_2$ and the academic 3D model case with four NN chains studied in Ref. 22 for the latter. A typical curve for a point in the triatic 3-MBS phase is shown in Fig. 3. One realizes that a rather weak critical IC of the order of few percents will remove the triatic phase in favor of the usual dipolar one becoming a "conic" or "umbrella" phase in external magnetic fields. The reduced saturation field h_s for the INC phase on the 1-magnon side is described by an exact expression obtained within spin wave theory (SWT):

$$h_s \equiv g\mu_{\rm B} H_{\rm s}/ \mid J_1 \mid = 2\alpha \left(1 - 0.25/\alpha\right)^2 + \tilde{j}_{\rm ic}, \qquad (1)$$

where $\tilde{j}_{\rm ic} = N_{\rm ic} j_{\rm ic}$ and $N_{\rm ic}$=2 or 4 for a 2D and a square-lattice chain arrangement, respectively. Using the Curie-Weiss temperature $\Theta_{\rm CW} = -0.5 \left[J_1 + J_2 + 0.5 N_{\rm ic} J_{\rm ic}\right]$ which determines the high-T ($T \gg -J_1, J_2$) spin susceptibility $\chi(T) \sim 1/(T - \Theta_{\rm CW})$, the IC entering Eq. (1) can be eliminated. As a result in the INC dipolar phase one obtains an expression convenient for the extraction of J_1 or α from H_s and $\Theta_{\rm CW}$:

$$G(J_{\rm ic}) = g\mu_{\rm B} H_{\rm s} + 4\Theta_{\rm CW} = \mid J_1 \mid \left[1 + 1/(8\alpha)\right]. \qquad (2)$$

Similar equations for $G = \mid J_1 \mid f(\alpha)$ can be derived also in other parameter regimes, where f is a relative simple function affected by the type of the IC and the region of the MP phase diagram. For instance in the cases of Li_2CuO_2 and $Ca_2Y_2Cu_5O_{10}$ to be considered below ignoring NNN IC and long range inchain coupling such as J_3 and J_4, we have simply $f = 2(1 - \alpha)$ (see Ref. 13, a generalization to include these very weak couplings, too is straightforward at the price of a more tedious f). Thus, these equations for G connect properties affected by the IC with single chain properties on the right hand. In most case it is much easier to determine the latter theoretically whereas the former can be measured experimentally.

The reduced form of $\theta = -2\Theta_{\rm CW}/J_1$ provides a useful constraints for the exchange parameters:

$$1 = \alpha + \beta + \gamma + \theta + (D - 1)\left[j_{\rm ic,\perp} + 2\sum_f j_{\rm ic,f}\right], \qquad (3)$$

where in addition to α further reduced inchain and IC quantities $\beta = -J_3/J_1$, $\gamma = -J_4/J_1$, $j_{\rm ic,\perp} = -J_{\rm ic,\perp}/J_1$ and analogously for nonperpendiculiar "frustrating" couplings have been introduced. $D = 3$ stands for a square-lattice chain arrangement. Applying Eq. (2) to Li_2ZrCuO_4 we predict a saturation field H_s of about 13 T using $J_1 = 273$ K, $\alpha = 0.3$, and $\Theta_{\rm CW} = 93$ K[8]. The small 1D-value of $H_{\rm s} \approx 4.5$ T, only, clearly demonstrates the importance of the IC in the vicinity of the FM-spiral critical point where $H_s \to 0$. The validity of Eq. (2) is guaranteed by a twice as large IC $j_{\rm ic} \sim 0.21$[9] compared with the critical value of 0.134 as deduced from Fig. 2. Thus, we are clearly outside the regions of triatic and the quartic phases and deep enough in the usual dipolar region. A similar 1-magnon picture holds for $Li(Na)Cu_2O_2$ where the critical IC coupling for a nematic phase is exceeded by a factor of 3. In fact, from the LDA derived IC and J_1 we estimate $j_{\rm ic} \approx 0.7$ well above 0.14 for the critical IC coupling strength taken from Fig. 2 for $\alpha \approx 1$ for $LiCu_2O_2$[25]. Similarly, using the empirical values for $NaCu_2O_2$: $\Theta_{\rm CW} = -41$ K taken from the $1/\chi(T)$ data between 350 and 400 K, $\alpha = 1.9$, $J_1 \approx -47.5$ K, and $g_a = 2.06$[27], we predict a saturation field H_s of about 155 T. From Eq. (1) we estimate $j_{\rm IC} \sim 0.2$ which clearly exceeds its critical value of about 0.12 (see Fig. 2). Using instead Eq. (2) and adopting a weak $J_3 \approx$5 K due to the dominant J_2 one estimates even 0.72 in accord with the LDA estimate given in Ref. 25. The difference between these estimates might be caused by the 1D approach used in Ref. 8 to extract J_1 and α in the presence of a sizable IC.

The inspection of the inset of Fig. 2 shows that the nematic 2-MBS phase region reaches a maximum as a

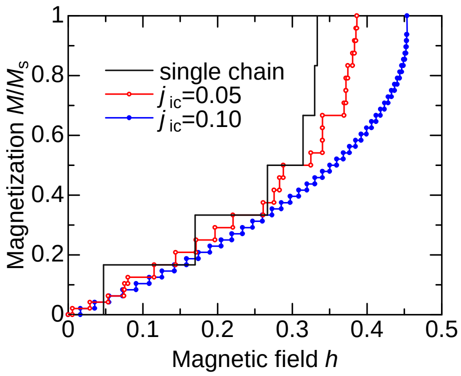

FIG. 3: (Color) Magnetization vs. applied external field for a 2D arrangement of unshifted chains with a direct AFM IC as modelled by four chains with $N = 24$ sites in each chain for different strengths of the IC $j_{\rm ic} \equiv J_{\rm ic}/ \mid J_1 \mid$ and an inchain frustration $\alpha = 0.5$ The criticaL value of $j_{\rm ic}$ at $h = h_{\rm s}$ amounts 0.088 for D=3 and 0.094 for $D = 2$.

function of the inchain frustration ratio $\alpha = -J_2/J_1$ at $\alpha \approx 1.103$. Noteworthy, its position and height (≈ 0.071688) differs somewhat from the feature shown in Fig. 6 of Ref. 22 : ≈ 0.62 and 0.057, respectively. We ascribe these differences to the field theory approximations used there (exact in the limit $\alpha \gg 1$).

B. Shifted frustrated chains in 2D and 3D

Let us now turn to the case of shifted chains with frustrated IC as in Li_2CuO_2 (see Figs. 1,4). If the IC is strong enough, MBS are supressed and only 1-magnon excitations survive at high fields. Then H_s reads

$$g\mu_{\rm B}H_{\rm s} = \tilde{J}_{\rm ic} + \tilde{J}'_{\rm ic} \text{ if } J_{ic} \geq J^{cr}_{ic,1}, \ \tilde{J}_{\rm ic} = N_{\rm ic}J_{\rm ic} \qquad (4)$$

where $N_{\rm ic}$ =8 denotes the number of interchain NN sites. Eq. (4) is valid for $j_{ic}(\alpha) > (4\alpha - 1)/9 \equiv j^{\rm cr}_{\rm ic,1}$ for $j'_{\rm ic} = 0$; $j_{\rm ic}(1/3) \approx 0.03704$. These parameters are very close to those for Li_2CuO_2: neglecting the exchange anisotropy, we obtained from INS data[12]: $J_{ic} \approx 9.04\text{K} > J^{\rm cr}_{\rm ic,1}(0.332) = -0.0364J_1 \approx 8.2$ K for $J'_{ic} = 0$. Then $H_{\rm s}$ depends *solely* on the IC. It can be directly read off from the extrapolated experimental value of $H_{\rm s}(T = 0)$[13]. For weaker IC h_s depends also somewhat on the intrachain parameter α. This has been clearly observed in our SWT and DMRG calculations. The latter also suggest that in this intermediate INC-phase above a second critical IC $j^{\rm cr}_{\rm ic,2} \approx 0.0109$ only specific INC 1-magnon low-energy excitations exist, see Fig. 4. Below $j^{\rm cr}_{\rm ic,2}$ 3-MBS are recovered as low-energy excitations and only this narrow region is 1D-like. The commensurate (C) phase behaves like an ordinary 3D antiferromagnet despite its seemingly quasi-1D nature. Finally, we note that for an unfrustrated AFM IC, the C-phase is missing. There is only one critical IC separating INC 1-magnon excitations from 3-MBS.

C. $LiVCuO_4$

From Eq. (3) a constraint for $LiVCuO_4$ follows. Indeed, from $1/\chi(T)$ at 500 K $\leq T \leq$ 650 K a small *FM* value $\Theta_{\rm CW} = 7.4$ K can be fitted from the data shown in Ref. 28 in contrast with "fits" employing the 300 K region resulting in artificial AFM values of ~ -25 to -15 K[29]. Thus, $\theta \leq 0.15$ holds for $-J_1 \geq 100$ K and α must be near 0.9±0.1 to get a weak IC. Thereby a weak diagonal IC of ∼ -10 K more or less close to that reported from INS data[28] and a third neighbor inchain coupling J_3 =-5 K have been adopted. The values $\alpha \sim 0.5$ to 0.6 and $J_1 \approx$ -180 K proposed in Ref. 16 are incompatible with a weak IC. A weak IC is suggested also by the weak magnetic moment at $T = 0$ due to strong quantum fluctuations being three times smaler than for Li_2CuO_2 and $Ca_2Y_2Cu_5O_{10}$ with frustrating AFM IC. We conclude, that for $\alpha \approx 0.9 \pm 0.1$ and $-J_1 \approx 100$ K nematics could be observed in $LiVCuO_4$. For a weak FM(AFM) IC the saturation field can be estimated using a fit of the numerically *exact* quasi 1D-solution[10]:

$$h_{\rm s}(\alpha) \ \approx \ h^{\rm 1D}_{\rm s}(\alpha) + \tilde{j}_{\rm ic}/2 + \eta(\alpha)j^2_{\rm ic}, \qquad (5)$$

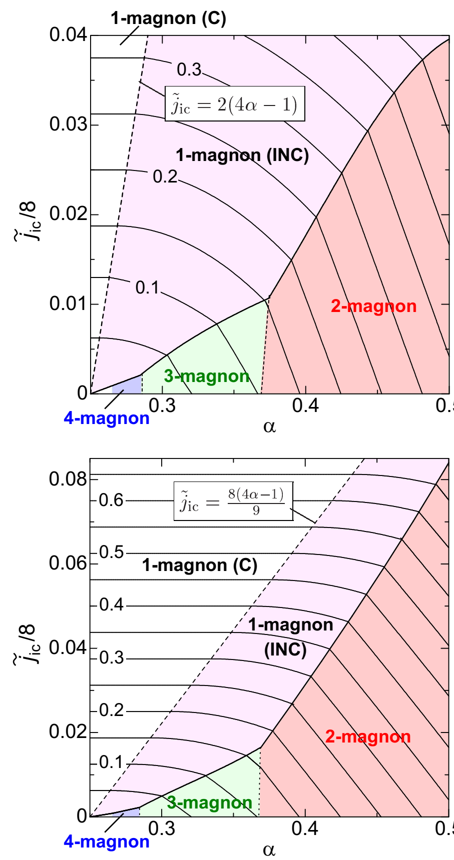

FIG. 4: (Color) 3D DMRG phase diagrams for frustrated IC . The saturation field h_s (given by contour lines) as a function of the reduced normalized IC $\tilde{j}_{\rm ic}$ and the inchain frustration α. Lower(Upper): (un)shifted chains with $\pm 3/2b$ ($\pm b$) IC.

$$h_s^{1D}(\alpha) \; = \; 2\alpha \left(1 - 0.25/\alpha^2 + 0.5/\alpha\right)/(1+1/\alpha), \quad (6)$$

where $\eta(\alpha) \approx 0.89 + 0.63084\alpha$ for $\alpha \leq 3$. The result is in accord with the data, if $-J_1$ is close to 100 K. Indeed, $J_1 = -98$ K, $\alpha = 0.83$, and $g = 2.13$ yield 55 T for $J_{ic,\perp}$ =8.8 K or 4.4 K expected for diagonal coupling to the NNN of the adjacent chain in a 2D arrangement of unshifted chains. For a FM IC of $J_{ic,\perp} = -9.9$ K or -5 K for a diagonal coupling a $J_1 = 94.8$ K can be found from Eq. (5). More advanced studies including explicitly the diagonal IC are necessary to refine all numbers. In particular, the quantum effect of J_{ic} on the pitch angle is of interest. A systematic study of FM IC as well as of the J_1-J_2-J_3 inchain model is without the scope of our paper and will be considered elsewhere. Now the challenging point is to find other MP exotic phases beyond the 2-magnon Bose condensation[21] triggered by the additional attractive FM IC.

V. SUMMARY AND CONCLUSIONS

To summarize, the crucial role of realistic AFM interchain coupling in quasi-1D helimagnets has been demonstrated. The rich and exotic physics of multipolar phases recently predicted for single chains is very sensitive to the strength of the IC. It can be readily eliminated by a weak AFM IC especially for triatic, quartic, etc. phases. For most CuO_2 chain systems studied so far, except probably $LiVCuO_4$, where a nematic phase or some other exotic phase might be expected, the AFM IC is too strong to allow for multipolar phases. The examination of weak FM IC as well as of anisotropy effects present in real materials is under study.

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2 MBS appear at ambient fields also in Ising-type anisotropic 2D systems, ladders and more complex systems. See e.g.: C.J. Hamer, Phys. Rev. B **79**, 212413 (2009), S. Dusuel *et al.*, Phys. Rev. B **81**, 064412 (2010) and references therein. Related aspects for frustrated chain cuprates will be considered elsewhere.

3 R. Bursill, G.A. Gehring, D.J.J. Farnell, J.B. Parkinson, T. Xiang, and C. Zheng J. Phys.: Condensed Matter **7** 8605 (1995).

4 T. Vekua *et al.*, Phys. Rev. B **76**, 174420 (2007).

5 F. Heidrich-Meisner *et al.*, Phys. Rev. B **74**, 020403R (2008).

6 S.-L. Drechsler *et al.*, J. Magnet. & Magnet. Mat. **290**, 345 (2005), –, ibid. **316** 306 (2007).

7 D. Dmitriev and V.Ya. Krivnov, Phys. Rev. B **79**, 054421 (2009).

8 S.-L. Drechsler *et al.*, Phys. Rev. Lett. **98**, 077202 (2007).

9 M. Schmitt, J. Mälek, S.-L. Drechsler, and H. Rosner, Phys. Rev. B **80**, 205111 (2009).

10 R. Kuzian and S.-L. Drechsler, Phys. Rev. B **75**, 024401 (2007).

11 R. Zinke, S.-L. Drechsler, and J. Richter, Phys. Rev. B **79**, 094425 (2009).

12 W.E.A. Lorenz, R.O. Kuzian, S.-L. Drechsler, W.D. Stein, N. Wizent, G. Behr, J. Málek, U. Nitzsche, H. Rosner, A. Hiess, W. Schmidt, R. Klingeler, M. Loewenhaupt, and B. Büchner, Europhys. Lett. **88**, 37002 (2009).

13 S. Nishimoto *et al.* arXiv:1004.3300 (2010).

14 A.V. Chubukov, Phys. Rev. B **44**, 4693 (1991).

15 M. Härtel J. Richter, D. Ihle, and S.-L. Drechsler, Phys. Rev. B ibid. **78**, 174412 (2008).

16 J. Sirker, Phys. Rev. B **81**, 014419 (2010).

17 L. Kecke, T. Momoi, and A. Furusaki, Phys. Rev. B, **76**, 060407 (2007).

18 T. Hikihara, L. Kecke, T. Momoi, and A. Furusaki, Phys. Rev. **78**, 144404 (2008).

19 J. Sudan, Lüscher, and A.M. Läuchli, Phys. Rev. B **80** 140402(R) (2009).

20 H. Katsura *et al.*, Phys. Rev. Lett. **101**, 187207 (2008).

21 M. Zhitomirsky *et al.* arXiv:1003.4096v1.

22 H.T. Ueda and K. Totsuka, Phys. Rev. B (2009).

23 S.R. White, Phys. Rev. Lett. **69**, 2863 (1992); Phys. Rev. B **48**, 10345 (1993).

24 S. Qin, S. Liang, Z. Su, and L. Yu, Phys. Rev. B **52**, R5475 (1995).

25 A.A. Gippius, E.N. Morozova, A.S. Moskvin, A,V. Zalessky, A.A. Bush, M. Baenitz, H. Rosner, and S.-L. Drechsler, Phys. Rev. B **70**, 020406(R) (2004).

26 T. Masuda *et al.*, Phys. Rev. B, **72**, 014405 (2005).

27 S.-L. Drechsler, J. Richter, A.A. Gippius, A. Vasiliev, A.A. Bush, A.S. Moskvin, J. Málek, Y. Prots, W. Schnelle, and H. Rosner, *et al.*, Europhys. Lett. **73**, 83 (2006).

28 M. Enderle, C. Mukherjee, B. Fak, R.K. Kremer, J.-M. Broto, H. Rosner, S.-L. Drechsler, J. Richter, J. Málek, A. Prokofiev, W. Assmu, S. Pujol, J.-L. Raggazzomi, H. Rakoto, M. Rheinstadter, and H.M. Ronnow, Europhys. Lett. **70**, 337 (2006).

29 A. Vasiliev *et al.*, Phys. Rev. B **64**, (2002).