

# Influence of interchain coupling on spiral ground-state correlations in frustrated spin- $\frac{1}{2}$ $J_1$ - $J_2$ Heisenberg chains

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We investigate the influence of an interchain coupling on the spiral ground-state correlations of a spin-1/2 Heisenberg model consisting of a two-dimensional array of coupled chains with nearest- ( $J_1$ ) and frustrating next-nearest-neighbor ( $J_2$ ) in-chain exchange couplings. Using the coupled cluster method we calculate the transition point between the commensurate and the incommensurate (spiral) ground states as well as the quantum pitch angle of the spiral ground state. In addition, we provide a simple empirical formula which describes the relation between the quantum pitch angle and the frustration parameter  $J_2/J_1$ .

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## I. INTRODUCTION

In recent years strongly frustrated quantum magnets exhibiting exotic ground-state (GS) phases have been intensively investigated both theoretically and experimentally (see, e.g., Refs. 1–3). At zero temperature all transitions between GS phases are driven purely by the interplay between quantum fluctuations and the competition between interactions (e.g., frustration) (see, e.g., Refs. 4–8). Particular attention has been paid to one-dimensional (1D)  $J_1$ - $J_2$  quantum Heisenberg models, which have been studied theoretically with much success over the last two decades (see Ref. 7 and references therein). From the experimental side recent investigations have shown that edge-shared chain cuprates build a special family of frustrated quantum magnets which can be described by a quasi-1D  $J_1$ - $J_2$  Heisenberg model. Among others we mention here  $\text{LiVCuO}_4$ ,  $\text{LiCu}_2\text{O}_2$ ,  $\text{NaCu}_2\text{O}_2$ ,  $\text{Li}_2\text{ZrCuO}_4$ , and  $\text{Li}_2\text{CuO}_2$ ,<sup>9–19</sup> which were identified as quasi-1D frustrated spin-1/2 magnets with ferromagnetic nearest-neighbor (NN) in-chain  $J_1$  and antiferromagnetic next-nearest-neighbor (NNN) in-chain interactions  $J_2$ . These cuprates have attracted much attention due to strong quantum effects and the observation of incommensurate spiral (helical) spin-spin correlations at low temperatures. Among these materials  $\text{Li}_2\text{ZrCuO}_4$  and  $\text{Li}_2\text{CuO}_2$  are of particular interest since these compounds are found to be near a quantum critical point.<sup>17,19</sup>

The 1D frustrated spin-1/2  $J_1$ - $J_2$  Heisenberg model may serve as the simplest model to describe some important features of such materials. The GS properties of the model in the classical limit are well known, i.e., when the spin quantum number  $s \rightarrow \infty$ . In this case the GS does exhibit a second-order transition from a collinear phase (ferro or antiferro) to a noncollinear phase with spiral correlations along the chains at  $J_2 = |J_1|/4$ . For  $J_2 \geq |J_1|/4$  the classical spiral (pitch) angle  $\alpha_{\text{cl}}$  is given by

$$\alpha_{\text{cl}} = \arccos(-J_1/4J_2). \quad (1)$$

Similar expressions can be derived in the presence of further couplings  $J_3$ ,  $J_4$ , etc.; however, this more general case will not be considered here for the sake of simplicity. Note that in the classical limit neither the pitch angle  $\alpha_{\text{cl}}$  nor the transi-

tion point  $J_2 = |J_1|/4$  depends on the interchain coupling  $J_\perp$ . Such a classical relation as Eq. (1) between  $J_1, J_2, \dots$  and the pitch angle  $\alpha_{\text{cl}}$  has been used to justify 1D parameter sets obtained from fitting  $\chi(T)$  data<sup>20</sup> or by local-density approximation (LDA) mapping procedures.<sup>21</sup> However, ignoring in this way sizable quantum effects, such an approach is not very convincing. Furthermore, in real materials additional terms in the Hamiltonian such as anisotropy or exchange coupling between the chains might be of relevance to yield a quantitative theoretical description of the experimental results. The existence of helical long-range order at low temperatures makes the importance of the interchain coupling evident. However, from the theoretical side such extended models so far are much less studied than the pure 1D “parent” models.

Therefore, in the present paper we focus on the discussion of the effect of the interchain coupling  $J_\perp$  on the GS spin-spin correlations in frustrated spin-1/2 Heisenberg model consisting of a two-dimensional (2D) array of frustrated chains coupled by  $J_\perp$  (see Fig. 1). In particular, we will discuss GSs with incommensurate spiral (i.e., noncollinear) correlations. Although, meanwhile many papers exist dealing with spiral correlations in the strictly 1D system ( $J_\perp = 0$ ),<sup>22–28</sup> the influence of the interchain coupling on the pitch angle to the best of our knowledge has not been discussed so far.

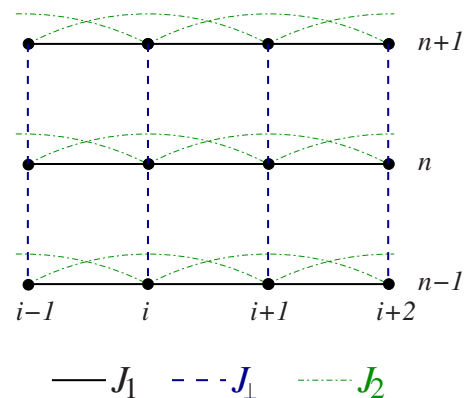


FIG. 1. (Color online) Illustration of the considered model [Eq. (2)] of coupled frustrated Heisenberg chains.

However, theoretical results for the considered system are highly desirable since meanwhile there is an increasing amount of experimental results for quasi-1D frustrated magnets with spiral correlations which need theoretical explanation.

The theoretical treatment of frustrated quantum antiferromagnets is far from being trivial. Although one can find exact GSs in some exceptional cases (see, e.g., Refs. 22 and 29–32) standard many-body methods may fail or become computational infeasible. For instance, the quantum Monte Carlo techniques suffer from the minus-sign problem in frustrated systems. The density-matrix renormalization group (DMRG) successfully used to discuss spiral correlations in 1D magnets<sup>23,25</sup> is essentially restricted to 1D systems, at least in the present state of the art. Also the exact diagonalization technique used in Ref. 26 to find the pitch angle for the 1D problem would be limited to extremely small chain lengths when a finite interchain coupling should be considered. Spin-wave theory, one of the most successful approximations for spin systems, starts from the classical GS. Within the linear spin-wave theory the classical pitch angle is not renormalized. A renormalization of the pitch angle can only be obtained by going beyond linear spin-wave theory in the  $1/S$  expansion, e.g., by using the Schwinger boson mean-field theory. Schwinger boson mean-field theory was used, e.g., in Ref. 33 for a similar (but not identical) system of coupled chains, namely, a frustrated coupled ladder system.

Another method fulfilling the requirement to be able to deal with frustrated spin systems at any dimension, including magnetic systems with incommensurate spiral GSs, is the coupled cluster method (CCM). This method was already used for the strictly 1D  $J_1$ - $J_2$  Heisenberg model and it was shown that the CCM results are in good agreement with the DMRG data.<sup>23</sup> Hence, in the present paper we use the CCM following the lines of Ref. 23 but extend the CCM calculations by including the interchain coupling  $J_\perp$ .

The frustrated spatially anisotropic 2D  $J_1$ - $J_2$ - $J_\perp$  spin-1/2-Heisenberg model (see Fig. 1) considered here reads

$$H = \sum_n \left\{ \sum_i [J_1 \mathbf{s}_{i,n} \cdot \mathbf{s}_{i+1,n} + J_2 \mathbf{s}_{i,n} \cdot \mathbf{s}_{i+2,n}] \right\} + \sum_i \sum_n J_\perp \mathbf{s}_{i,n} \cdot \mathbf{s}_{i,n+1}, \quad (2)$$

where the index  $n$  labels the chains and  $i$  labels the lattice sites within a chain  $n$ . The NN in-chain coupling  $J_1$  is fixed to either  $J_1=1$  (antiferromagnetic) or  $J_1=-1$  (ferromagnetic). The interchain coupling  $J_\perp$  and the frustrating NNN in-chain coupling  $J_2$  are considered as varying parameters of the model. Note that in the case considered here the interchain coupling does not lead to frustration and it is practically of arbitrary strength, i.e., including also the region beyond the quasi-1D limit  $|J_\perp| \ll |J_1|, J_2$ . Anyhow, the opposite limit  $|J_\perp| \gg |J_1|, J_2$  will not be considered for reasons of lacking physical relevance (to the best of our knowledge). We focus on the extreme quantum case, i.e.,  $(\mathbf{s}_{i,n})^2 = s(s+1)$  with  $s=1/2$ . The main point which will be considered here is the influence of the interchain coupling  $J_\perp$  on the transition point between the collinear phase and the noncollinear spiral phase

and on the pitch angle characterizing the spiral correlations in the quantum model with  $s=1/2$ .

## II. COUPLED CLUSTER METHOD

In this section we outline only some main features of the CCM which are relevant for the model under consideration. Again we mention that we follow the lines described in Ref. 23 where the CCM was applied to the strictly 1D problem. For more details of the method the interested reader is referred to Refs. 23 and 34–46. Special attention to the CCM treatment of noncollinear GSs was paid in Refs. 23, 39–41, 43, and 44.

The starting point for the CCM calculation is the choice of a normalized reference or model state  $|\Phi\rangle$ , together with a complete set of (mutually commuting) multiconfigurational creation operators  $\{C_L^+\}$  and the corresponding set of their Hermitian adjoints  $\{C_L\}$ ,

$$\langle \Phi | C_L^+ = 0 = C_L | \Phi \rangle, \quad \forall L \neq 0, \quad C_0^+ \equiv 1, \quad (3)$$

$$[C_L^+, C_K^+] = 0 = [C_L, C_K]. \quad (4)$$

With the set  $\{|\Phi\rangle, C_L^+\}$  the CCM parametrization of the ket GS eigenvector  $|\Psi\rangle$  of the considered many-body system is then given by

$$|\Psi\rangle = e^S |\Phi\rangle, \quad S = \sum_{L \neq 0} a_L C_L^+. \quad (5)$$

The CCM correlation operator  $S$  contains the correlation coefficients  $a_L$  which can be determined by the so-called set of ket equations

$$\langle \Phi | C_L e^{-S} H e^S | \Phi \rangle = 0, \quad \forall L \neq 0. \quad (6)$$

For a more detailed discussion of the basic features of the CCM we refer to Ref. 36. Here we give only some more details of the method which are specific for the considered frustrated quantum spin system. We choose a reference state corresponding to the classical state of the spin model, i.e., the ferromagnetic state  $|\downarrow\downarrow\downarrow\downarrow\downarrow\cdots\rangle$  along a chain for  $J_1=-1$  and small  $J_2$  and the Néel state  $|\downarrow\uparrow\downarrow\uparrow\cdots\rangle$  for  $J_1=1$  and small  $J_2$ , whereas for larger frustration  $J_2$  we have to choose a noncollinear reference state with spiral correlations along the chains (i.e., pictorially,  $|\Phi\rangle = |\uparrow \nearrow \rightarrow \searrow \downarrow \swarrow \cdots\rangle$ ) characterized by a pitch angle  $\alpha$ , i.e.,  $|\Phi\rangle = |\Phi(\alpha)\rangle$ . Such states include the ferromagnetic state ( $\alpha=0$ ) as well as the Néel state ( $\alpha=\pi$ ). In the quantum model the pitch angle may be different from the corresponding classical value  $\alpha_{cl}$ . Hence, we do not choose the classical result for the pitch angle but, rather, we consider  $\alpha$  as a free parameter in the CCM calculation, which has to be determined by minimization of the GS energy given in the CCM formalism by  $E(\alpha) = \langle \Phi(\alpha) | e^{-S} H e^S | \Phi(\alpha) \rangle$ , i.e., from  $dE/d\alpha|_{\alpha=\alpha_{qu}} = 0$  [see also the Appendix, Eqs. (A1)–(A6)].

In order to find an appropriate set of creation operators it is convenient to perform a rotation of the local axes of each of the spins, such that all spins in the reference state align in the negative  $z$  direction. This rotation by an appropriate local angle  $\delta_{i,n}$  of the spin on lattice site  $(i, n)$  is equivalent to the spin-operator transformation

$$\left. \begin{aligned} s_{i,n}^x &= \cos \delta_{i,n} \hat{s}_{i,n}^x + \sin \delta_{i,n} \hat{s}_{i,n}^z, & s_{i,n}^y &= \hat{s}_{i,n}^y \\ s_{i,n}^z &= -\sin \delta_{i,n} \hat{s}_{i,n}^x + \cos \delta_{i,n} \hat{s}_{i,n}^z \end{aligned} \right\}. \quad (7)$$

The local rotation angle  $\delta_{i,n}$  can be easily expressed by the pitch angle  $\alpha$  of the spiral reference state, where the relation  $\delta_{i,n}(\alpha)$  depends on the signs of  $J_1, J_\perp$  and the lattice vector  $\mathbf{R}_{i,n}$ .

In this rotated set of local spin coordinates the reference state and the corresponding creation operators  $C_L^+$  are given by

$$|\hat{\Phi}\rangle = |\downarrow\downarrow\downarrow\downarrow\cdots\rangle, \quad C_L^+ = \hat{s}_{i,n}^+, \hat{s}_{i,n}^+, \hat{s}_{j,m}^+, \hat{s}_{i,n}^+, \hat{s}_{j,m}^+, \hat{s}_{k,l}^+, \dots, \quad (8)$$

where the indices  $(i,n), (j,m), (k,l), \dots$  denote arbitrary lattice sites. This specified form of the creation operators  $C_L^+$  and the corresponding reference state  $|\hat{\Phi}\rangle$  immediately make clear that the general relations (3) and (4) are fulfilled. Moreover, it is also obvious that for spin-half systems one has  $(C_L^+)^n = 0$  for  $n > 1$ . In the rotated coordinate frame the Hamiltonian (2) becomes dependent on the pitch angle  $\alpha$ . It reads

$$\begin{aligned} H = & \frac{J_1}{4} \sum_{i,n} [\cos(\alpha) + 1] (\hat{s}_{i,n}^+ \hat{s}_{i+1,n}^- + \hat{s}_{i,n}^- \hat{s}_{i+1,n}^+) + [\cos(\alpha) - 1] \\ & \times (\hat{s}_{i,n}^+ \hat{s}_{i+1,n}^+ + \hat{s}_{i,n}^- \hat{s}_{i+1,n}^-) + 2 \sin(\alpha) [\hat{s}_{i,n}^+ \hat{s}_{i+1,n}^z - \hat{s}_{i,n}^- \hat{s}_{i+1,n}^z \\ & + \hat{s}_{i,n}^z \hat{s}_{i+1,n}^z - \hat{s}_{i,n}^- \hat{s}_{i+1,n}^z] + 4 \cos(\alpha) \hat{s}_{i,n}^z \hat{s}_{i+1,n}^z \\ & + \frac{J_2}{4} \sum_{i,n} [\cos(2\alpha) + 1] (\hat{s}_{i,n}^+ \hat{s}_{i+2,n}^- + \hat{s}_{i,n}^- \hat{s}_{i+2,n}^+) + [\cos(2\alpha) - 1] \\ & \times (\hat{s}_{i,n}^+ \hat{s}_{i+2,n}^+ + \hat{s}_{i,n}^- \hat{s}_{i+2,n}^-) + 2 \sin(2\alpha) [\hat{s}_{i,n}^+ \hat{s}_{i+2,n}^z - \hat{s}_{i,n}^- \hat{s}_{i+2,n}^z \\ & + \hat{s}_{i,n}^z \hat{s}_{i+2,n}^z - \hat{s}_{i,n}^- \hat{s}_{i+2,n}^z] + 4 \cos(2\alpha) \hat{s}_{i,n}^z \hat{s}_{i+2,n}^z \\ & - \frac{J_\perp^A}{2} \sum_{i,n} (\hat{s}_{i,n}^+ \hat{s}_{i,n+1}^+ + \hat{s}_{i,n}^- \hat{s}_{i,n+1}^- + 2 \hat{s}_{i,n}^z \hat{s}_{i,n+1}^z) \\ & + \frac{J_\perp^F}{2} \sum_{i,n} (\hat{s}_{i,n}^+ \hat{s}_{i,n+1}^- + \hat{s}_{i,n}^- \hat{s}_{i,n+1}^+ + 2 \hat{s}_{i,n}^z \hat{s}_{i,n+1}^z), \end{aligned} \quad (9)$$

where  $\hat{s}_{i,n}^\pm \equiv \hat{s}_{i,n}^x \pm i \hat{s}_{i,n}^y$  and  $\alpha (\equiv \delta_{i,n} - \delta_{i+1,n})$  is the pitch angle between the two neighboring spins in a chain interacting via the NN bond  $J_1$ , which has to be determined for the quantum model. For ferromagnetically coupled spin chains ( $J_\perp^F \neq 0$ ) one has to set  $J_\perp^A = 0$  in Eq. (9) and vice versa. Therefore, from Eq. (9) it is obvious that in the quantum case considered here the role of the (unfrustrated) interchain coupling  $J_\perp$  introduced in Eq. (2) is different for ferromagnetic and antiferromagnetic  $J_\perp$ . By contrast, in the classical case a corresponding interchain coupling does not affect the pitch angle at all.

The CCM formalism would be exact if we could take into account all possible multispin configurations in the correlation operator  $S$  which is impossible to do in practice for a quantum many-body system. Hence, it is necessary to truncate the expansions of  $S$ . In Ref. 23 it was demonstrated that the so-called SUB2-3 approximation for the strictly 1D system leads to results of comparable accuracy to those obtained using the DMRG method. In the SUB2-3 approximation all

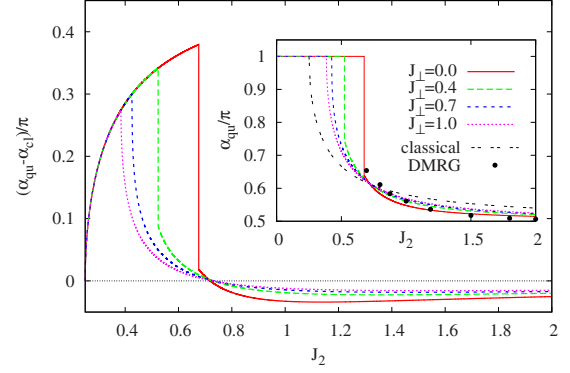


FIG. 2. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_2$  for antiferromagnetic  $J_1$  and  $J_\perp$  [case (i)]. The main panel shows the difference between the quantum pitch angle  $\alpha_{\text{qu}}$  and its classical counterpart  $\alpha_{\text{cl}}$  for various interchain couplings  $J_\perp$ . The inset shows the corresponding data for the quantum pitch angle  $\alpha_{\text{qu}}$  itself. For comparison the classical pitch angle and the DMRG data of White and Affleck (Ref. 25) for the strictly 1D quantum model are also shown.

configurations are included which span a range of no more than three contiguous sites and contain only two or fewer spins. A particular advantage of the SUB2-3 approximation consists of the possibility to find the relevant CCM equation (6) in closed analytical form (see the Appendix). These explicit equations provided here can be used to find the quantum pitch angle  $\alpha_{\text{qu}}$  for an arbitrary set of parameters  $J_1, J_2$ , and  $J_\perp$  by simple numerical solution of them.

### III. RESULTS

For the model under consideration we have calculated that point  $J_2^S$  where the GS state spin-spin correlations change from collinear to noncollinear spiral correlations as well as the quantum pitch angle  $\alpha_{\text{qu}}$ . In what follows we call the point  $J_2^S$  the “transition point.” We mention, however, that the question for magnetic GS long-range order goes beyond the scope of the present paper. Generally one can argue that for the strictly 1D problem the GS (except the simple ferromagnetic state) does not exhibit magnetic long-range order, whereas for finite  $J_\perp$  GS long-range order can exist (cf., e.g., Refs. 47–51). In particular, the GS phase for larger  $J_2$  and small  $J_\perp$  is magnetically disordered and may have a weak spontaneous dimerization along with finite-range incommensurate magnetic correlations.<sup>47</sup> Hence the transition point  $J_2^S$  for small  $J_\perp$  may locate that narrow parameter region, where finite-range magnetic correlations are changing from commensurate to incommensurate ones, but does not indicate a true quantum phase transition.

We present data for  $J_\perp = \pm 0, 0.1, 0.2, \dots, 1.0$  and a fine net of  $J_2$  values. For the sake of clarity in the following we assort the results into four cases, depending on the signs of  $J_1$  and  $J_\perp$ , namely, (i)  $J_1 = 1$  and  $J_\perp \geq 0$ , (ii)  $J_1 = 1$  and  $J_\perp \leq 0$ , (iii)  $J_1 = -1$  and  $J_\perp \geq 0$ , and (iv)  $J_1 = -1$  and  $J_\perp \leq 0$ .

Since for cases (i) and (ii) the behavior is quite similar, we can discuss both cases together. The pitch angle in dependence on  $J_2$  is shown in Fig. 2 [case (i)] and Fig. 3 [case (ii)].

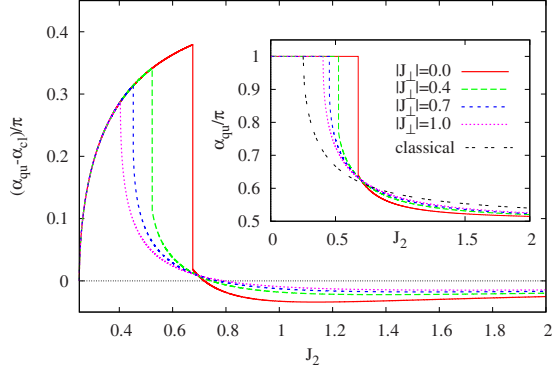


FIG. 3. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_2$  for antiferromagnetic  $J_1$  and ferromagnetic  $J_{\perp}$  [case (ii)]. The main panel shows the difference between the quantum pitch angle  $\alpha_{\text{qu}}$  and its classical counterpart  $\alpha_{\text{cl}}$  for various interchain couplings  $J_{\perp}$ . The inset shows the corresponding data for the quantum pitch angle  $\alpha_{\text{qu}}$  itself. For comparison the classical pitch angle is also shown.

For comparison we also draw the corresponding DMRG data of White and Affleck<sup>25</sup> for the strictly 1D problem in Fig. 2. These data agree quite well with the CCM data in particular for larger  $J_2$ . It is obvious that quantum fluctuations change the classical correlations drastically. In particular, in contrast to the classical case, the collinear quantum state can survive into the region  $J_2 > J_2^{\text{cl}} = 0.25$ , where classically it is already unstable. This effect is known as *order from disorder*<sup>52,53</sup> and is widely observed in quantum spin systems.<sup>39,43,54,55</sup> We find, e.g.,  $J_2^s \approx 0.68$  for the quantum model with  $J_{\perp} = 0$ , which is in good agreement with known results (see, e.g., Fig. 3 in Ref. 26).

Switching on  $J_{\perp}$ , i.e., increasing the dimension of the spin system, the effect of quantum fluctuations should become weaker and, as a result, the collinear quantum state gives way for the state with incommensurate correlations at smaller values of  $J_2$ , i.e., the transition point moves toward the classical value (see Fig. 4). However, even in nearly isotropic 2D systems ( $J_{\perp} \sim 1$ ) the quantum fluctuations are still important and one has  $J_2^s \approx 0.38$  ( $J_2^s \approx 0.40$ ) for case (i) [for case (ii)] which is significantly above the classical value.

The shift of the transition point leads to an interesting behavior of the difference  $\alpha_{\text{qu}} - \alpha_{\text{cl}}$ . It is positive for 0.25

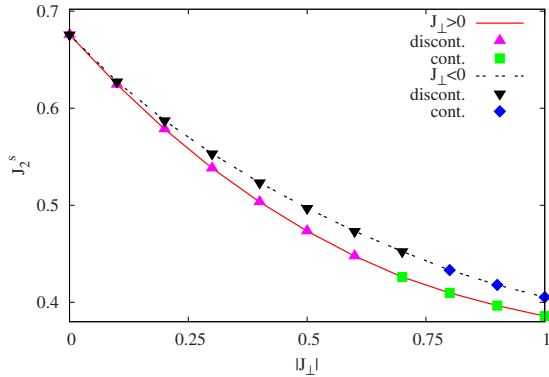


FIG. 4. (Color online) The transition point  $J_2^s$  as a function of the interchain coupling  $J_{\perp}$  for  $J_1 = 1$  and  $J_{\perp} > 0$  [case (i)] as well as  $J_{\perp} < 0$  [case (ii)]. The triangles indicate a discontinuous and the squares a continuous change of  $\alpha_{\text{qu}}$ .

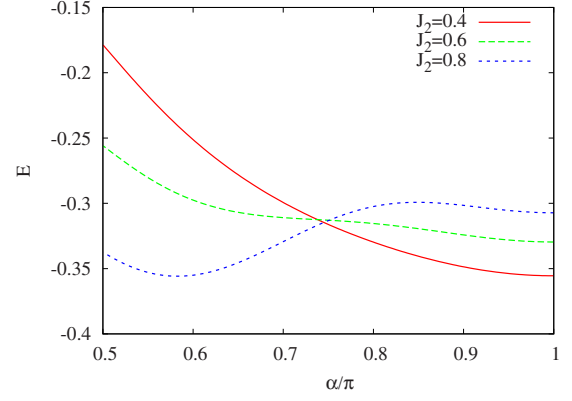


FIG. 5. (Color online) The GS energy versus pitch angle  $\alpha$  for different (fixed)  $J_2$  and antiferromagnetic  $J_1$  ( $J_{\perp} = 0$ ).

$< J_2 < J_2^*$  but negative for  $J_2 > J_2^*$ . Surprisingly we find  $J_2^* \approx 0.7$  as almost independent of  $J_{\perp}$ , i.e., all curves  $\alpha_{\text{qu}} - \alpha_{\text{cl}}$  vs  $J_2$  meet approximately in one point (see Figs. 2 and 3). Note that for large  $J_2 > J_1$  the model with strongest quantum fluctuations, i.e.,  $J_{\perp} = 0$ , approaches the limit  $\alpha_{\text{qu}} \rightarrow \pi/2$  most rapidly.

Next we discuss the behavior of the pitch angle at the transition point. For smaller values of the interchain coupling [ $J_{\perp} \leq 0.7$  for case (i) and  $J_{\perp} \leq 0.8$  for case (ii)] we find a discontinuous behavior of the quantum pitch angle. A similar jumpwise change of the pitch angle has been found for 2D frustrated quantum spin models.<sup>39,43</sup> Note that in Ref. 23 the CCM curve was terminated before the jump occurs, so that the jump has not been observed there.<sup>56</sup> The discontinuous change of  $\alpha_{\text{qu}}$  is related to the existence of two minima in the  $E$  versus  $\alpha$  curve and takes place for parameter values where both minima have equal depth (cf. Fig. 5). For larger  $J_{\perp}$  (weaker quantum fluctuations)  $E(\alpha)$  exhibits only one minimum and the quantum pitch angle is changing continuously from the collinear to the spiral GS. We remind the reader that for the classical case the transition from the commensurate to the incommensurate GS takes place at  $J_2 = |J_1|/4$  and that it is continuous for any value of  $J_{\perp}$ .

Finally, in Fig. 6 we have drawn the pitch angle  $\alpha_{\text{qu}}$  in dependence on the interchain coupling  $J_{\perp}$  for case (i). As

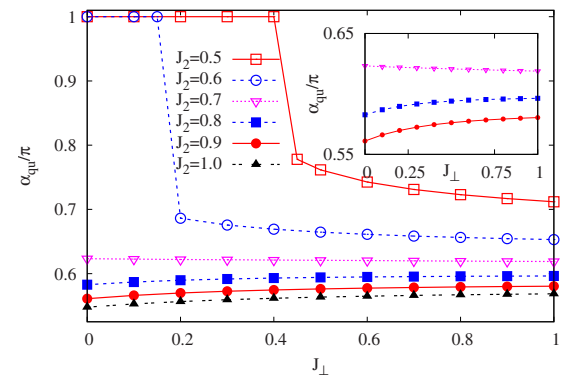


FIG. 6. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_{\perp}$  for antiferromagnetic  $J_1 = 1$  [case (i)] and various values of the frustrating NNN exchange  $J_2$  (the lines are guides for the eyes). The inset shows the data for  $J_2 = 0.7, 0.8, 0.9$  with an enlarged scale.

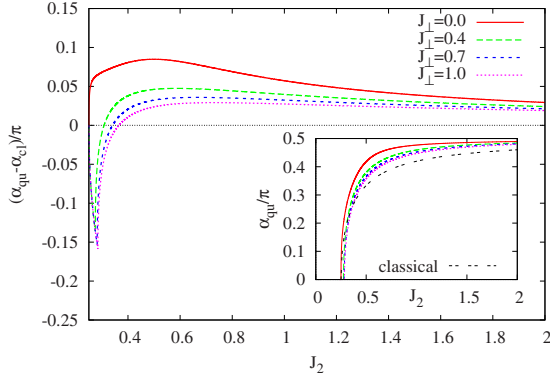


FIG. 7. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_2$  for ferromagnetic  $J_1$  and antiferromagnetic  $J_{\perp}$  [case (iii)]. The main panel shows the difference between the quantum pitch angle  $\alpha_{\text{qu}}$  and its classical counterpart  $\alpha_{\text{cl}}$  for various interchain couplings  $J_{\perp}$ . The inset shows the corresponding data for the quantum pitch angle  $\alpha_{\text{qu}}$  itself. For comparison the classical pitch angle is also shown.

discussed above, with increasing  $J_{\perp}$  the quantum pitch angle moves toward the corresponding classical value. However, in accordance with the above discussion of the change in the sign of  $\alpha_{\text{qu}} - \alpha_{\text{cl}}$  we find two different regimes: for  $J_2 \leq J_2^*$  the  $\alpha_{\text{qu}}$  increases with growing  $J_{\perp}$  while for  $J_2 \geq J_2^*$  the  $\alpha_{\text{qu}}$  decreases with  $J_{\perp}$ .

Let us now pass to cases (iii) and (iv), i.e., when  $J_1 = -1$  is ferromagnetic. In contrast to cases (i) and (ii), we find that here the transition from the collinear state to the spiral state is always continuous. The reason for that can be again attributed to the strength of quantum fluctuations. For ferromagnetic  $J_1$  the relevant collinear state at  $J_2 < J_2^s$  consists of ferromagnetic chains, having a classical (i.e., “nonfluctuating”) GS, coupled by  $J_{\perp}$ . While for case (iv) the ferromagnetic interchain coupling does not change this nonfluctuating GS at all, for case (iii) due to the presence of antiferromagnetic couplings quantum fluctuations become relevant and the GS becomes a true quantum state, but the change in magnetic correlations within the chains for  $J_{\perp} > 0$  remains weak. Hence, virtually no [case (iv)] or only weak [case (iii)] quantum fluctuations occur at the transition from the collinear to the noncollinear GS. As a result the transition takes place precisely at  $J_2 = |J_1|/4$  for case (iv) while for case (iii) the transition point  $J_2^s$  is above the classical value indicating again an order from disorder effect. However, the shift of  $J_2^s$  is small because the quantum fluctuations are weak (see Figs. 7 and 9). Nevertheless, such a small shift of  $J_2^s$  due to a finite interchain coupling might be important for systems such as  $\text{Li}_2\text{ZrCuO}_4$  and  $\text{Li}_2\text{CuO}_2$  (Refs. 17 and 19) which are near the transition point. Note that Bader and Schilling<sup>57</sup> first found that the transition point is fixed at  $J_2 = |J_1|/4$ , if the collinear state is the classical ferromagnetic one. Note further that a similar change from a discontinuous to a continuous transition was discussed in Ref. 40.

The quantum pitch angle for cases (iii) and (iv) is shown in Figs. 7 and 8. Obviously, there is also a significant difference between the quantum and the classical pitch angles; however, it is smaller than for cases (i) and (ii). For  $J_{\perp} = 0$  the largest difference of about  $0.09\pi$  is found at  $J_2 \approx 0.5$ . For

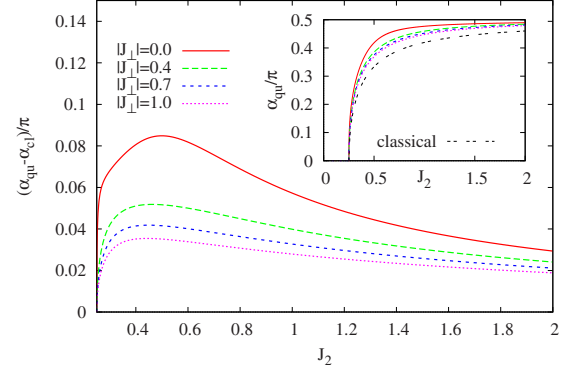


FIG. 8. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_2$  for ferromagnetic  $J_1$  and  $J_{\perp}$  [case (iv)]. The main panel shows the difference between the quantum pitch angle  $\alpha_{\text{qu}}$  and its classical counterpart  $\alpha_{\text{cl}}$  for various interchain couplings  $J_{\perp}$ . The inset shows the corresponding data for the quantum pitch angle  $\alpha_{\text{qu}}$  itself. For comparison the classical pitch angle is also shown.

case (iv) for all values of  $J_{\perp} \leq 0$  and  $J_2 > 0.25$  the quantum pitch angle is larger than the classical one. On the other hand, for case (iii) the shift of the transition point  $J_2^s$  leads to a change in the sign of  $\alpha_{\text{qu}} - \alpha_{\text{cl}}$ , i.e., the antiferromagnetic interchain coupling yields a more subtle change in the GS correlations by quantum fluctuations. Moreover, due to the shift of  $J_2^s$  we find a quite large difference  $|\alpha_{\text{qu}} - \alpha_{\text{cl}}|$  for  $J_{\perp} > 0$  and  $J_2 \approx 0.27 - 0.28$  (see Figs. 7 and 9).

Similarly for cases (i) and (ii) we observe that for large  $J_2 > J_1$  the model with strongest quantum fluctuations, i.e.,  $J_{\perp} = 0$ , approaches the limit  $\alpha_{\text{qu}} \rightarrow \pi/2$  most rapidly. We mention here that a large value of  $|J_2/J_1|$  is realized, e.g., in  $\text{LiCuVO}_4$ ,<sup>9,12,15</sup> for which  $J_2/J_1 \approx -2.4$  has been estimated.<sup>12,15</sup> The variation of  $\alpha_{\text{qu}}$  with  $J_{\perp}$  for various  $J_2$  is shown in Fig. 10. It can be seen that the variation of  $\alpha_{\text{qu}}$  with  $J_{\perp}$  is largest for small  $J_{\perp}$ . Furthermore, in difference to cases (i) and (ii)  $\alpha_{\text{qu}}$  decreases monotonously with  $J_{\perp}$  for all values of  $J_2 > J_2^s$ .

As mentioned in Sec. I, ignoring quantum effects a classical relation like Eq. (1) between the exchange couplings and the pitch angle has been used to discuss the  $J_2/J_1$  ratio in Refs. 20 and 21. To overcome this problem we will provide an empirical formula that fits the continuous part of  $\alpha_{\text{qu}}(J_2)$

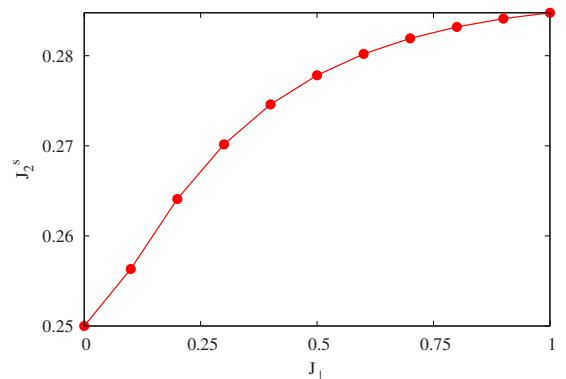


FIG. 9. (Color online) The transition point  $J_2^s$  as a function of the interchain coupling  $J_{\perp}$  for  $J_1 = -1$  and  $J_{\perp} > 0$  [case (iii)].

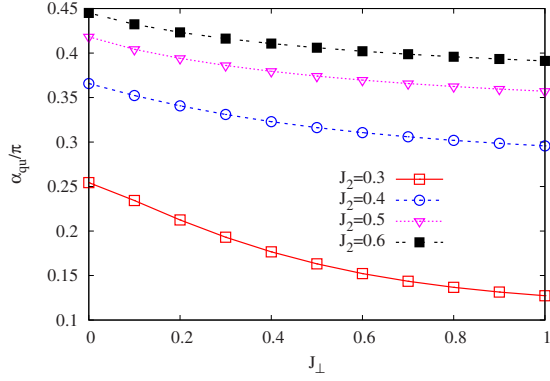


FIG. 10. (Color online) The quantum pitch angle  $\alpha_{\text{qu}}$  versus  $J_{\perp}$  for ferromagnetic  $J_1 = -1$  [case (iii)] and various values of the frustrating NNN exchange  $J_2$ .

given in Figs. 2, 3, 7, and 8 very well. Having in mind that the shape of the continuous part of  $\alpha_{\text{qu}}(J_2)$  resembles the classical behavior, we find that  $\alpha_{\text{qu}}(J_2)$  is well approximated by (written now in dimensional exchange units for the convenience of application in real experimental situations)

$$\alpha_{\text{qu}}(J_2) = \arccos \left\{ \frac{-J_1}{\left[4(J_2 - J_2^s + \frac{1}{4}|J_1|)\right]^{\nu}} \right\} \quad (10)$$

with the exponent  $\nu$  as fitting parameter. Obviously, Eqs. (1) and (10) coincide for  $J_2^s = |J_1|/4$  and  $\nu = 1$ . In Fig. 11 we show  $\nu$  in dependence on  $J_{\perp}$  for the four cases (i)–(iv). We find that  $\nu$  is always larger than the classical value  $\nu_{\text{cl}} = 1$ . In accordance with the above discussion,  $\nu$  decreases with increasing  $J_{\perp}$ ; i.e., it goes toward the classical exponent  $\nu_{\text{cl}}$ . From the experimental point of view the edge-shared chain cuprates are of particular interest. The parameter situation of these compounds corresponds to case (iii). Hence, we give here simple fit formulas for that case which describe the behavior of  $J_2^s(J_{\perp})$  as shown in Fig. 9 and the behavior of  $\nu(J_{\perp})$  as shown in Fig. 11 by the red line. We find that the relation

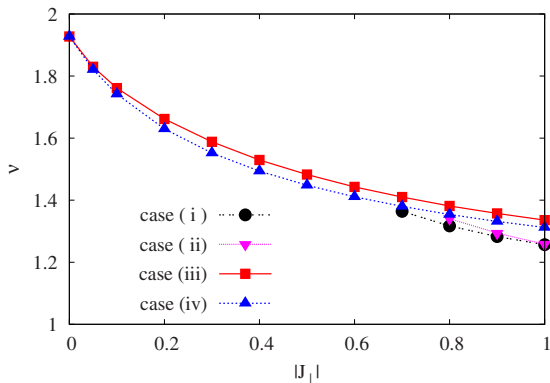


FIG. 11. (Color online) The exponent  $\nu$  [see Eq. (10)] in dependence on  $J_{\perp}$  for the cases (i)–(iv).

TABLE I. Numerical values for the exponent  $\nu$  and the transition point  $J_2^s$  in dependence on  $J_{\perp}$  for the cases (i)–(iv) [cf. Eq. (10)].

$ J_{\perp} $	(i): $J_1 = 1, J_{\perp} > 0$		(ii): $J_1 = 1, J_{\perp} < 0$	
	$J_2^s$	$\nu$	$J_2^s$	$\nu$
0.70	0.426	1.364		
0.80	0.409	1.317	0.433	1.341
0.90	0.397	1.283	0.418	1.294
1.00	0.386	1.256	0.405	1.258
$ J_{\perp} $	(iii): $J_1 = -1, J_{\perp} > 0$		(iv): $J_1 = -1, J_{\perp} < 0$	
	$J_2^s$	$\nu$	$J_2^s$	$\nu$
0.00	0.250	1.972	0.250	1.972
0.10	0.256	1.761	0.250	1.742
0.20	0.264	1.662	0.250	1.630
0.30	0.270	1.588	0.250	1.552
0.40	0.275	1.530	0.250	1.494
0.50	0.278	1.483	0.250	1.448
0.60	0.280	1.443	0.250	1.411
0.70	0.282	1.410	0.250	1.380
0.80	0.283	1.381	0.250	1.354
0.90	0.284	1.357	0.250	1.331
1.00	0.285	1.336	0.250	1.312

$$\nu = \frac{a}{\left(\frac{J_{\perp}}{|J_1|} + b\right)^c}, \quad J_2^s = \frac{|J_1|}{4} + p|J_1| \tanh\left(q \frac{J_{\perp}}{|J_1|}\right) \quad (11)$$

with  $a = 1.37$ ,  $b = 0.13$ ,  $c = 0.17$ ,  $p = 0.036$ , and  $q = 2.11$  provide a reasonable fit of our data for case (iii). In addition, we present numbers for  $J_2^s$  and  $\nu$  for various values of  $J_{\perp}$  for all cases in Table I. The fitting formula (11) as well as the data in Table I can be used in combination with Eq. (10) to fix the  $J_2/J_1$  ratio using the pitch angle  $\alpha$  as an input, e.g., from neutron scattering.<sup>9,10,12,20</sup>

Finally, we note that recently the pitch angle of  $\text{Li}_2\text{ZrCuO}_4$  has been determined from  $^7\text{Li}$ -NMR data to amount  $\alpha = 33^\circ \pm 2^\circ$ .<sup>18</sup> This value corresponds to a predicted ratio  $-J_2/J_1 = 0.298$  within the framework of the classical Eq. (1). This value is surprisingly very close to the ratio  $-J_2/J_1 = 0.3$  estimated from thermodynamic properties within the 1D-quantum spin-1/2  $J_1$ - $J_2$  model.<sup>17</sup> However, from Fig. 10 for a realistic weak effective antiferromagnetic interchain coupling  $J_{\perp} \leq 0.1|J_1|$  a somewhat larger pitch angle would be predicted, namely,  $\alpha \approx 42^\circ$  for  $-J_2/J_1 = 0.3$ . Hence, other factors such as a sizeable exchange anisotropy are expected to be relevant in this material. Such an anisotropy may lead to a modification of the classical spiral as well as to a reduction in quantum fluctuation compared with the isotropic-spin model considered here. The corresponding effects are outside the scope of the present paper. They will be considered elsewhere.

#### IV. SUMMARY

Based on the CCM we have studied the GS correlations of a 2D array of frustrated spin-1/2  $J_1$ - $J_2$  chains coupled by an interchain exchange interaction  $J_\perp$ . We have discussed the influence of quantum fluctuations, frustration, and interchain coupling on the pitch angle and the transition point between a GS with collinear commensurate correlations and a GS with incommensurate spiral correlations. Using the CCM within the so-called SUB2-3 approximation we obtain a closed set of analytical equations which can be used to calculate the pitch angle for an arbitrary set of exchange parameters. We have found that for  $J_2 > 0.25|J_1|$  the pitch angle of the quantum model significantly deviates from the classical value and can be strongly influenced by the interchain coupling  $J_\perp$ . Furthermore, we have observed that the quantum pitch angle approaches its limiting value of  $\pi/2$  for increasing values of  $J_2$  much faster than for the classical model. For several combinations of the sign of  $J_1$  and  $J_\perp$  we have found that the change in the pitch angle within the quantum model could be discontinuous while the change of the pitch angle within the classical model is always continuous.

The problem of antiferromagnetically or ferromagnetically coupled frustrated spin-1/2  $J_1$ - $J_2$  chains investigated in detail in this paper to the best of our knowledge so far has not been considered. Hence, it would be interesting to compare our CCM results with those to be obtained by other methods such as the Schwinger boson approach which is an alternative method to deal with noncollinear ground states in 2D quantum spin systems.<sup>33</sup>

#### ACKNOWLEDGMENTS

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#### APPENDIX: CCM SUB2-3 APPROXIMATION

Within the SUB2-3 approximation scheme the CCM correlation operator  $S$  [see Eq. (5)] contains five nonequivalent correlation coefficients  $a_L$  ( $L=1, \dots, 5$ ) corresponding to the lattice configurations shown in Fig. 12. These configurations (or lattice animals) represent the arrangement of spin operators acting on the lattice spins. To each configuration belongs a corresponding ket equation [see Eq. (6)]. The set of these ket equations can be found by a bit tedious but straightforward calculation. It reads

$$\begin{aligned} & \frac{J_1}{4} \{ [\cos(\alpha) - 1] (1 - 12a_1^2 + 8a_2^2 + 8a_3^2 + 8a_4^2 + 24a_5^2) \\ & + 4a_2 [\cos(\alpha) + 1] - 8a_1 \cos(\alpha) \} + \frac{J_2}{4} \{ [\cos(2\alpha) - 1] \\ & \times (16a_3a_5 - 16a_1a_2) + 4a_1 [\cos(2\alpha) + 1] \\ & - 16a_1 \cos(2\alpha) \} + \frac{J_\perp^F}{2} (8a_5 - 8a_1) \\ & - \frac{J_\perp^A}{2} (16a_4a_5 - 16a_1a_3 + 16a_2a_5 - 8a_1) = 0, \quad (\text{A1}) \end{aligned}$$

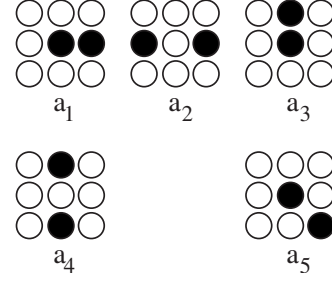


FIG. 12. Illustration of the five configurations on the spin lattice which contribute to the CCM SUB2-3 approximation. Each configuration is related to a particular multiconfigurational creation operators  $\{C_L^\dagger\}$  and to the corresponding correlation coefficient  $a_L$  ( $L=1, \dots, 5$ ) [see Eqs. (5) and (8)]. The circles in the figure represent lattice sites; the black circles indicate the position of the flipped spins in a certain configuration.

$$\begin{aligned} & \frac{J_1}{4} \{ [\cos(\alpha) - 1] (-16a_1a_2 + 16a_3a_5) + 4a_1 [\cos(\alpha) + 1] \\ & - 16a_2 \cos(\alpha) \} + \frac{J_2}{4} \{ [\cos(2\alpha) - 1] (1 - 12a_2^2 + 8a_1^2 + 8a_3^2 \\ & + 8a_4^2 + 16a_5^2) - 8a_2 \cos(2\alpha) \} - \frac{J_\perp^F}{2} (8a_2) \\ & - \frac{J_\perp^A}{2} (16a_1a_5 - 16a_2a_3 - 8a_2) = 0, \quad (\text{A2}) \end{aligned}$$

$$\begin{aligned} & \frac{J_1}{4} \{ [\cos(\alpha) - 1] (-16a_1a_3 + 16a_2a_5 + 16a_4a_5) \\ & + 8a_5 [\cos(\alpha) + 1] - 16a_3 \cos(\alpha) \} + \frac{J_2}{4} \{ [\cos(2\alpha) - 1] \\ & \times (-16a_2a_3 + 16a_1a_5) - 16a_3 \cos(2\alpha) \} + \frac{J_\perp^F}{2} (4a_4 - 4a_3) \\ & - \frac{J_\perp^A}{2} (1 - 12a_3^2 + 8a_1^2 + 8a_2^2 + 8a_4^2 + 24a_5^2 - 4a_3) = 0, \quad (\text{A3}) \end{aligned}$$

$$\begin{aligned} & \frac{J_1}{4} \{ [\cos(\alpha) - 1] (-16a_1a_4 + 16a_3a_5) - 16a_4 \cos(\alpha) \} \\ & + \frac{J_2}{4} \{ [\cos(2\alpha) - 1] (-16a_2a_4 + 8a_5^2) - 16a_4 \cos(2\alpha) \} \\ & + \frac{J_\perp^F}{2} (4a_3 - 8a_4) - \frac{J_\perp^A}{2} (-16a_3a_4 + 16a_1a_5 - 8a_4) = 0, \quad (\text{A4}) \end{aligned}$$

$$\begin{aligned} & \frac{J_1}{4} \{ [\cos(\alpha) - 1] (16a_2a_3 - 16a_1a_5 + 16a_3a_4) \\ & + 8a_3 [\cos(\alpha) + 1] - 32a_5 \cos(\alpha) \} + \frac{J_2}{4} \{ [\cos(2\alpha) - 1] \\ & \times (16a_1a_3 - 32a_2a_5 + 16a_4a_5) + 8a_5 [\cos(2\alpha) + 1] \end{aligned}$$

$$-32a_5 \cos(2\alpha)\} + \frac{J_{\perp}^F}{2}(8a_1 - 16a_5) - \frac{J_{\perp}^A}{2} \\ \times (16a_1a_2 - 16a_3a_5 + 16a_1a_4 - 16a_5) = 0. \quad (\text{A5})$$

The GS energy is a function of (some of) these correlation coefficients and of the pitch angle  $\alpha$ . It reads

$$E = \frac{J_1}{4}\{2a_1[\cos(\alpha) - 1] + \cos(\alpha)\} + \frac{J_2}{4}\{2a_2[\cos(2\alpha) - 1] \\ + \cos(2\alpha)\} + \frac{J_{\perp}^F}{4} - \frac{J_{\perp}^A}{4}(4a_3 + 1). \quad (\text{A6})$$

Note that in the above equations one has to set  $J_{\perp}^A$

$= 0$  ( $J_{\perp}^F = 0$ ) and to replace  $J_{\perp}^F$  with  $J_{\perp}$  ( $J_{\perp}^A$  with  $J_{\perp}$ ) for ferromagnetic (antiferromagnetic)  $J_{\perp}$ . To determine the quantum pitch angle  $\alpha_{\text{qu}}$  as a function of the parameters  $J_1$ ,  $J_2$ , and  $J_{\perp}$ , one has to solve the equation  $dE/d\alpha|_{\alpha=\alpha_{\text{qu}}} = 0$  (cf. Sec. II) together with the set of ket equations (A1)–(A5) self-consistently by standard numerics.

Finally, we will illustrate some limiting cases contained in Eqs. (A1)–(A5). For  $J_{\perp} = 0$  one has  $a_3 = a_4 = a_5 = 0$  and the remaining two nontrivial Eqs. (A1) and (A2) then coincide with the corresponding equations given in Ref. 23. In case of  $J_2 = 0$ ,  $J_{\perp} = 0$ , and  $a_3 = a_4 = a_5 = 0$  but  $J_1 \neq 0$  (or alternatively,  $J_2 = 0$ ,  $J_1 = 0$ , and  $a_1 = a_2 = a_5 = 0$  but  $J_{\perp} \neq 0$ ) one finds the two ket equations for the simple unfrustrated linear chain.

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