Field-Induced Deformation of Magnetic Microphases in Frustrated Ising Magnets

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The frustrated triangular Ising magnet Ca$_3$Co$_2$O$_6$ has long been known to exhibit peculiar slow spin dynamics in various forms, such as the evenly-spaced metastable magnetization steps and the long-wavelength spin density wave order. While the extraordinarily long timescale of spin relaxation impedes previous experimental studies in equilibrium, a recently-proposed elaborate field-cooling protocol has opened up a possibility of investigating the magnetic phase diagram much closer to equilibrium. Motivated by this progress, we combine Monte Carlo simulations and Ginzburg-Landau theory to study the magnetic field-induced deformation of the spin density wave order. We also discuss previously reported experimental phase diagrams, likely out of equilibrium.

Frustrated spin systems can reveal exotic physics due to quasi-degenerate states [1]. Even a classical system can realize novel spin textures and quasiparticles, such as skyrmion crystals [2, 3], spin ice and monopoles [4–6], and soliton structures in the axial next-nearest-neighbor Ising (ANNNI) model [7, 8]. Their sensitivity to perturbations makes them attractive not only as potential devices in some cases, but also as platforms to study far-from-equilibrium dynamics due to metastable states [5, 9].

Ever since the late 90’s, Ca$_3$Co$_2$O$_6$ (CCO) has earned considerable attention for its extremely slow spin dynamics and other unusual magnetic behaviors [10–13]. In CCO, trigonal prismatic Co$^{3+}$ $S = 2$ sites form ferromagnetic (FM) Ising chains running along the $c$ axis, while arranged in a triangular lattice in the $ab$ plane coupled by weak antiferromagnetic (AFM) interactions (Fig. 1) [14]. Below spin-freezing temperature $T_{SF} \approx 5$ K, striking evenly-spaced metamagnetization steps appear under a magnetic field sweep [12], which has been the subject of a long-time debate [12, 13, 15–19]. Interestingly, many characteristics such as $T_{SF}$ and step heights are sensitive to measurements, e.g., the sweep rate, with the only exception of the evenly-spaced transition fields $\approx 1.2$ T, 2.4 T, and 3.6 T with additional steps at higher fields [16]. Some theory invoked an analogy with quantum tunneling in molecular-based magnets [17]. Meanwhile, frustration-induced, far-from-equilibrium nature of this phenomenon was pointed out by replacing each FM chain by an effective one-dimensional lattice used in the MF approximation, where only the interactions connected to the central site are shown. (c) Effective one-dimensional lattice used in the MF approximation, where an arrow represents a magnetic moment of a given $ab$ plane.

In this Letter, we discuss equilibrium in-field behaviors of CCO at low temperature. Although it has been notoriously difficult to access them experimentally, they nevertheless hold the key to understanding the peculiar slow dynamics and out-of-equilibrium behavior. In fact, the nature of the slow dynamics in CCO is more intricate than simply revealed by the metamagnetization steps. Resonant x-ray [26, 27] and neutron spectroscopies [18, 28–31] showed that CCO develops an incommensurate spin density wave (SDW) order below $T_{SDW} \approx 25$ K. The SDW order accompanies short-range order [28] and undergoes a low-$T$ order-order transition under slow cooling [32]. Unlike an earlier proposal of the partially-disordered AFM (PDA) state [12], the SDW state has a long-wavelength modulation along the $c$ axis, which coexists with other phases in a magnetic field [27, 31]. Firstly, these observations render the rigid chain interpretation ambiguous. More importantly, the modulation wavelength $\lambda_{SDW} \sim 10^3$ Å ($\approx 10^2$ sites) increases as temperature $T$ is lowered. The corresponding relaxation time also grows substantially, so that the system deviates from equilibrium already below $T \lesssim 13$ K [18], somewhat higher than $T_{SF}$. It was pointed out that the observed SDW state is essentially a soliton lattice as in the ANNNI model [33], a prototypical model for spontaneous superstructures due to competition between nearest and next nearest neighbor Ising interactions in one direction of a square or cubic lattice [7, 8]. The $T$-dependent change of $\lambda_{SDW}$ corresponds to different magnetic microphases [33], similar to other self-organizing modulated phases in physical and chemical systems [34]. Thus, CCO provides a rare intersection where the ANNNI model phenomenology meets out-of-equilibrium physics in a solid state system.

Furthermore, it was recently demonstrated that the slow spin dynamics can be bypassed by an elaborate field-cooling protocol, where every in-field measurement is performed af-
ter a separate cooling in the target magnetic field [35]. Remarkably, the protocol allowed for reaching the equilibrium 1/3 magnetization plateau down to \( T = 2 \) K without suffered from metastable states [35]. When combined with neutron and other spectroscopies, it has a potential for investigating field-induced deformation of the SDW state and incommensurate-commensurate (IC-C) transitions. The purpose of this work is to provide a theoretical guide for such future experiments by presenting an equilibrium in-field phase diagram for a realistic three-dimensional (3D) lattice model for CCO. We address both model-specific and universal physics by combining mean-field (MF) theory, MC simulations, and Ginzburg-Landau (GL) theory.

In CCO, \( S = 2 \) spins have large easy-axis anisotropy [12], which permits a description by an effective Ising model with \( \sigma_i^z = \pm 1 \),

\[
\hat{H} = \sum_{\nu=1,2,3} \sum_{\langle i,j \rangle_\nu} J_{ij} \sigma_i^z \sigma_j^z - h \sum_i \sigma_i^z, \tag{1}
\]

where \( h = g \mu_B S H \) with \( g, \mu_B \), and \( H \) being the g-factor, the Bohr magneton, and a magnetic field, respectively, and \( \langle i,j \rangle_\nu \) denotes neighboring sites connected by \( J_{ij}, \nu \in \{1,2,3\} \). Here, \( J_1 < 0 \) is the intrachain FM interaction and \( J_2 (J_3) \) is the AFM interchain interaction shifted by 1/3 (2/3) lattice parameters along the \( c \) axis (Fig. 1). An \textit{ab initio} study [36] suggested \( |J_1| \gg J_2 \approx J_3 \) and an NMR experiment reported \( J_1 = -23.9(2) \) K and \( J_2 + J_3 = 2.3(2) \) K, suggesting \( J_2 = 1.1 \) K and \( J_3 = 1.2 \) K to explain the SDW ordering vector [37]. Below, for simplicity, we assume \( J_2 = J_3 \).

Despite the apparent structural differences, CCO has the same kind of geometrical frustration as the ANNNI model due to the vertical shifts of the interchain interactions. After a few steps along a spiral path, \( J_2 \) and \( J_3 \) compete with \( J_1 \) [38]. We first consider a zero-field MF theory by assuming a FM order in each \( ab \) plane (separated by 1/3 lattice parameters from each other), as shown in Fig. 1(c). By extending the previous analysis [33], we obtain a phase diagram as a function of \( T \) and \( \kappa = J_2/|J_1| = J_3/|J_1| \) [Fig. 2(a)]. Below \( T_{\text{SDW}}(\kappa) \), the ordering wavevector \( \mathbf{Q} = (0,0,Q_3) \) varies quasi-continuously except for the low-\( T \) region below a lock-in IC-C transition \( T_{\text{IC-C}} \), where the MF solutions are \( \uparrow \uparrow \uparrow \) or \( \downarrow \downarrow \downarrow \) for \( \kappa < 1 \) and \( \uparrow \downarrow \downarrow \uparrow \) for \( \kappa > 1 \). A few extended commensurate regions exist as enclosed by thin lines in Fig. 2(a), though they are restricted to \( \kappa \approx 1 \) and therefore irrelevant in the application to CCO. The quasi-continuous change of \( Q_3 \) of \( (\kappa,T) \), which dominates the overall phase diagram, corresponds to numerous soliton lattice microphases [33].

Next, to demonstrate the ANNNI-like physics in an unbiased way, we run MC simulations, considering a lattice of size \( L \times L \times L_c \) with periodic boundary conditions (\( N_{\text{spin}} = 3L^2L_c \) spins). We fix \( \kappa = 0.1 \) and \( L/L_c = 40 \) in most cases below [39]. The investigated system sizes are \( 2 \times 2 \times 80–8 \times 8 \times 320 \). We combine single-spin and intra-chain cluster updates [33] with replica exchanges [40] performed every 10 MC steps. Several hundreds of replicas are needed for largest lattices (e.g., 400 replicas for \( 8 \times 8 \times 320 \) for \( \kappa \leq 0.2|J_1| \)) to maintain a reasonable exchange acceptance rate to guarantee efficient sampling at low temperatures.

The possible \( T \)-dependent change of the ordering wavevector \( \mathbf{Q} \) is a challenge in numerically investigating SDW states [41–43]. For a reliable determination of \( T_{\text{SDW}} \), we define the effective ordering wavevector \( \mathbf{Q}_{L}^{\text{eff}}(T) = (0,0,Q_{L}^{\text{eff}}) \) for each system size as one for the maximum of the spin structure factor \( S_{\mathbf{q}} = N_{\text{spin}}(|\langle M_{\mathbf{q}} \rangle^2| - |\langle M_{\mathbf{q}} \rangle|^2) \) with \( M_{\mathbf{q}} = N_{\text{spin}}^{-1} \sum_i \sigma_i^x \exp(-i \mathbf{q} \cdot \mathbf{r}_i) \), which is also defined for \( T > T_{\text{SDW}} \). Since \( \lim_{T \to \infty} Q_{L}^{\text{eff}}(T) = Q(T) \) is expected, we evaluate the Binder parameter \( U Q_{L}^{\text{eff}}(T) = \langle |M_{\mathbf{q}}^2(T)|^4 \rangle / \langle M_{\mathbf{q}}^4(T) \rangle^2 \) defined for the \( \mathbf{q} = Q_{L}^{\text{eff}}(T) \) component of the order parameter. We investigate the crossing points for different system sizes to evaluate \( T_{\text{SDW}} \) [39], as in the normal usage of the Binder parameter [44]. We also determine \( T_{\text{IC-C}} \) for the lock-in IC-C transition, or \( T_c \) for a direct transition, into a commensurate phase by using the Binder parameter [39].

The phase diagram for \( \kappa = 0.1 \) based on the MC data for \( T \approx 0.45|J_1| \) is shown in Fig. 2(b). In zero field, we obtain \( T_{\text{SDW}}/|J_1| = 1.407(5) \) and \( T_{\text{IC-C}}/|J_1| = 0.52(1) \). For \( h = 0 \), \( T_{\text{IC-C}} \) increases rapidly while \( T_{\text{SDW}} \) is almost constant in \( h \). Consequently, the SDW phase shrinks rapidly with increasing \( h \) and is confined in a relatively low-field regime \( h \leq 0.2|J| \approx 0.17h_{\text{sat}} \), where \( h_{\text{sat}} = 6(J_2 + J_3) \) is the saturation field. The SDW order realized in the model is sinusoidal [28], rather than square-wave-like [31], throughout the entire SDW phase. Above the field-induced multicritical (Lifshitz) point \( h_{\text{L}} \approx 0.2|J_1| \), there occurs a direct first-order transition into the ferrimagnetic (FIM) \( \uparrow \downarrow \uparrow \uparrow \) state, which forms the 1/3 magnetization plateau [35]. The discontinuous nature of the direct transition is consistent with the \( Z_2 \) symmetry breaking in \( d = 3 \), as in the three-state Potts model [45, 46].

The estimated \( T_{\text{SDW}} \) roughly coincides with the highest-\( T \) peak in the specific heat (Fig. 3). We also observe that \( Q_{L}^{\text{eff}} = (0,0,Q_{L}^{\text{eff}}) \) at \( T = T_{\text{SDW}} \) slightly but clearly deviates from \( q_{\text{com}} = (0,0,2\pi) \) [47] and varies towards \( q_{\text{com}} \) as further low-
ering $T$ below $T_{\text{SDW}}$ (Fig. 3). The observed step-like behavior of $Q_{\text{eff}}^0$, which also causes spurious peaks in $C$ at strongly size-dependent temperatures, is due to the finite-size discretization of the wavevector ($\Delta Q_{\text{eff}}^0/(2\pi) = 0.003125$ for $L_x = 320$). Considering the range of our system sizes, the most natural interpretation is that the change of $Q(T) = \lim_{L \to \infty} Q_{\text{eff}}^0(T)$ is (quasi-)continuous towards $q_{\text{com}}$. Another observation is that $Q(T \approx T_{\text{SDW}})$ is almost independent of $h$, with a possible deviation near the Lifshitz point [Fig. 3(c)]. The result implies that the larger the external magnetic field, the more rapid the $T$-dependent drift of $Q$ (or $\lambda_{\text{SDW}}$) in the SDW state, as shown in Fig. 3.

At low temperatures, the ordering wavevector is pinned at $q_{\text{com}} = (0, 0, 2\pi)$, corresponding to a three-sublattice order with restored translational symmetry along the $c$ axis. For $T \geq 0.45|J_1|$, we find no evidence of an additional order-order transition as reported in CCO [32]. The lock-in temperature $T_{\text{IC-C}}$, where $Q_{\text{eff}}^0(T) = 2\pi$ is reached upon decreasing $T$, agrees well with one estimated by using the Binder parameter (Fig. 3). The two main candidates for the three-sublattice order for $T < T_{\text{IC-C}}$ are the FIM state and the PDA state, similar to the case in other triangular lattice AFM Ising models [48–53]. A convenient indicator for finite-size calculations is $C_6 = \langle M_6^0 \rangle / \langle M_0 \rangle$, which takes $C_6 > 0$ ($C_6 < 0$) for the FIM (PDA) state [50]. For $h = 0$, we confirm the PDA state below $T_{\text{IC-C}}$, which, however, should be distinguished from the previous claim of the same state in CCO below $\approx 25$ K [12]. For $h \neq 0$, we find the FIM state at low $T$ down to $h/|J_1| = 0.025$, implying that the observed PDA state is extremely fragile against the magnetic field.

Finally, for a more universal description of commensurate and incommensurate phases in CCO and similar materials, such as Ca$_3$Co$_2$Mn$_2$O$_6$ [19, 54], Sr$_2$Ca$_2$CoMn$_2$O$_6$ [55], and Ca$_3$CoRhO$_6$ [56], we consider a GL theory in the long-wavelength limit. As we demonstrate below, the GL theory is in the same form as one for the ANNNI model [57, 58]. Moreover, the GL theory for the in-field ANNNI-like models [59] has yet been fully discussed despite its experimental relevance. By using the Hubbard-Stratonovich transformation, we introduce a complex order parameter $\phi(\mathbf{r}) \sim e^{i\Phi(\mathbf{r})}$ describing a coarse-grained local three-sublattice order [48], where $q_{\text{com}} = (0, 0, 2\pi)$ is a choice motivated by our MC results. Due to $J_2$ and $J_3$, the Fourier transform $J(\mathbf{q})$ of exchange interactions has minima at incommensurate wavevectors $\pm q_{\text{com}}$ with $q_{\text{min}} = (0, 0, 2\pi + \epsilon)$. The estimate for CCO is $\epsilon/(2\pi) = -0.0061$ for $J_2 = J_3 = 0.046|J_1|$ suggested by NMR [37]. On one hand, because of this deviation, a commensurate three-sublattice ordered state ($\phi(\mathbf{r}) \sim \text{const.}$) cannot acquire the full energy gain of the exchange interaction. On the other hand, such an unmodulated commensurate ordered state can have an additional contribution due to umklapp terms. Hence, the key role is played by the competition between gradient and umklapp terms favoring incommensuration and commensuration, respectively, as in the classic ANNNI model [58].

For $h = 0$, we find that the leading umklapp term is of the sixth order (the smallest even number satisfying $n q_{\text{com}} = (0, 0, 0)$ is $n = 6$):

$$\mathcal{H}_{h=0} = \int d^3r \left[ \frac{1}{2} \nabla \cdot \left( \nabla - i \hat{\epsilon} \right) \phi(\mathbf{r}) \right]^2 + t|\phi(\mathbf{r})|^2 + u_6|\phi(\mathbf{r})|^6 + u_4|\phi(\mathbf{r})|^4 + v_6(\phi(\mathbf{r})^6 + \phi^*(\mathbf{r})^6) \right].$$

where $t$, $u_4$, $u_6$, and $v_6$ are GL coefficients and $\hat{\epsilon} = (0, 0, 1)$. The effective GL Hamiltonian includes a gradient term that acts as adding a momentum $\Delta q = (0, 0, \epsilon)$ to $\phi$. At $T = T_{\text{SDW}}$, critical fluctuations renders $\phi(\mathbf{r})$ nonzero with this additional momentum $\Delta q$. The resulting state, $\phi(\mathbf{r}) \sim \text{(const.)} \times e^{i\Delta q \mathbf{r}}$, indeed corresponds to the three-sublattice long-wavelength SDW state with $Q = q_{\text{min}}$. The sixth-order $v_6$ term has no effect for this modulated mode at $T = T_{\text{SDW}}$; it is also irrelevant in the renormalization group sense, implying emergent $U(1)$ symmetry. The transition breaks translation symmetry.
along the c axis and will be in the 3D $XY$ universality class. The observed main peak of the specific heat exhibits a sign of smearing [Fig. 3(a)], which is consistent with the negative exponent $\alpha = -0.0146(8) < 0$ [60] for this universality class.

For $T < T_{SDW}$, the competition against the $v_6$ term sets in, which affects the phase factor of $\phi$, thereby $Q(T)$. We write $\phi(r) = A(r) e^{iQ(r)}$ and apply a MF decoupling for the massive amplitude fluctuation (“Higgs”) mode $\delta_6(r) = A(r) - (A)$ and the phase mode $\theta(r)$. We thereby obtain a sine-Gordon model for $\theta(r)$,

$$\mathcal{H}_{h=0,6} = (A)^2 \int d^3 r \left[ \frac{1}{2} (\nabla \theta(r) - e \hat{e})^2 + 2(A)^4 v_6 \cos 6\theta(r) \right].$$

(3)

The gradient term tends to drift the phase, which can lead to a plethora of soliton landscapes through the competition against the cosine term [58, 61], as demonstrated in our MF and MC studies (Fig. 2). As $T < T_{SDW}$ is lowered, $(A)$ grows, and the relative strength of the cosine term is enhanced. Consequently, the model is expected to undergo a lock-in transition. For $v_6 > 0$ ($v_6 < 0$), the phase is locked-in at $\theta = 2n\pi/6$ [9 $\theta = (2n + 1)\pi/6$] with an integer $0 \leq n < 6$, corresponding to the FIM (↑↑↓ or ↓↓↑) state and the PDA state [48–53], respectively. Although our MF calculation implies $v_6 > 0$, our unbiased MC simulation suggests $v_6 < 0$ in the present model in zero field.

For $h \neq 0$, the field-induced uniform $q = 0$ component $m(r)$ is allowed by symmetry. Consequently, additional lower-order umklapp terms can appear. In the present case, we find

$$\Delta \mathcal{H}_{0-q_m} \approx \int d^3 r w_4 m(r) \left( \phi(r)^3 + \phi^*(r)^3 \right),$$

(4)

as the leading-order contribution with a new coupling constant $w_4$. The total effective GL Hamiltonian for $h \neq 0$ is $\mathcal{H}_{h0,\phi,m} = \mathcal{H}_{h0,\phi} + \mathcal{H}_{q_m,0} + \Delta \mathcal{H}_{0-q_m}$, where $\mathcal{H}_{q_m,0} = \int d^3 r \left[ \frac{1}{2} (c \nabla m(r))^2 + \mu^2 m(r)^2 - hm(r) \right]$ is the noninteracting part for $m(r)$ with $c > 0$ and $\mu^2 > 0$ being the gapped spin wave parameters near $q = 0$. By a similar MF decoupling as above, we find

$$\Delta \mathcal{H}_{0-q_m,\theta} = 2w_4 \langle m \rangle A^3 \int d^3 r \cos 3\theta(r).$$

(5)

Firstly, only a subset of the FIM states are favored by $\Delta \mathcal{H}_{0-q_m,\theta}$ because of the reduced symmetry. Depending on the sign of $w_4$, the favored states are either ↑↑↓ or ↓↓↑, each of which is three-fold degenerate, though ↑↑↓ is naturally anticipated for $h > 0$. Secondly, since $\Delta \mathcal{H}_{0-q_m,\theta} \propto \langle m \rangle$, the strength of the field-induced umklapp term is enhanced by the magnetic field. Moreover, the prefactor is $\propto (A)^3$ as opposed to $\propto (A)^6$ in zero field, thus growing faster for $T < T_{SDW}$ in a magnetic field. Therefore, the region of the incommensurate SDW microphases is expected to be narrower for larger $h$, in excellent agreement with our MC results (Fig. 2).

Thirdly, when the zero-field umklapp term favors the PDA state, as found in the current model (1), the zero and field-induced umklapp terms ($\sim \cos \theta, \cos 6\theta$, respectively) compete against each other. This opens a possibility of a mixed phase below $T_{IC-C}$, though our MC results imply that the corresponding magnetic field region is, if any, extremely small. Finally, emergent $U(1)$ symmetry at $T = T_{SDW}$ is expected also for $h \neq 0$, because the additional momentum $\Delta q$ cancels the net contribution of the umklapp term.

In summary, we presented a comprehensive theory for magnetic field-induced effects on the equilibrium state of CCO and similar materials, such as $\text{Ca}_3\text{Co}_2\text{O}_6$ [19, 54], $\text{Sr}_3\text{Ca}_2\text{Co}_{2}\text{Mn}_6\text{O}_{19}$ [55], and $\text{Ca}_3\text{CoRhO}_6$ [56]. In a magnetic field, the deformation of SDW microphases as a function of $T$, as characterized by $Q(T)$, occurs much more rapidly, resulting in “steeper” Devil’s staircases than in zero field (Fig. 3). The deformation eventually leads to the IC-C transition into the PDA (FIM) state for $h = 0$ ($h \neq 0$) with a possibility of an unconfirmed mixed phase in an extremely low-field regime. Theoretically, these field-induced phenomena emerge through an indirect control of phase-locking terms in the effective sine-Gordon model by magnetic field and temperature.

Interestingly, the obtained SDW-FIM phase boundary nearly coincides with the boundary detected by $\mu$SR measurements [62] and also with one detected by weak anomaly in the magnetic entropy change [63]. A preliminary resonant x-ray study also reported a field-induced IC-C transition [27], though this experiment performed at 5 K likely reflects out-of-equilibrium phenomena at low temperatures. However, when combined with the recently-proposed elaborate field-cooling protocol [35], neutron scattering and other spectroscopies focusing on the low-field regime are promising to examine the field-induced behaviors discussed in this work, which may provide further insights in the peculiar slow dynamics and out-of-equilibrium behaviors in CCO and related materials.

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**Supplementary Material: 1. Aspect ratio in MC simulations**

The system undergoes the SDW transition as $T$ is lowered in the low-field regime. As a guiding principle for the aspect ratio of the system size in the MC simulation to study the SDW phase and the SDW phase transition, we demand that the lattice can host the modulation at $T = T_{SDW}$ with as little finite-size tension as possible. The ordering wavevector $Q$ at $T = T_{SDW}$ corresponds to the wavevectors $\pm q_{\text{min}}$ for the minima of $\langle Q \rangle$. For $J_3 \approx J_1 \approx 0.046|J_1|$ suggested by NMR [37], the minima are located at $\pm q_{\text{min}}/2\pi \approx \pm(0, 0.9939)$. Because $q_{\text{min}}$, and thus $Q$, is in this case very close to the three-sublattice commensurate wavevector $q_{\text{com}}$, even a single periodicity of the spin modulation requires a large number of unit
cells along the $c$ axis, $L_c = 2\pi/|q_{\text{min},3} - q_{\text{com},3}| \approx 160$ (three sites per unit cell). To ease this difficulty, we instead choose $J_2 = J_3 = 0.1|J_1|$, for which $q_{\text{min}}/2\pi \approx (0, 0, 0.987)$ and $\approx 80$ cells along the $c$ axis are needed for a single periodicity. As demonstrated for a very similar model studied in Ref. 33, with the only difference being in a transverse field, the model for $J_2 = J_3 = 0.1|J_1|$ also realize the long-wavelength SDW order, and therefore, essentially the same physics can be expected as in the case of even smaller values for $J_2$ and $J_3$. In this study, we set $L_c/L = 40$ and the investigated system sizes are $2 \times 2 \times 80$ to $8 \times 8 \times 320$.

**Supplementary Material: 2. Determination of $T_{SDW}$**

For a second-order phase transition into a commensurate ordering, say, with a momentum $\mathbf{q} = \mathbf{Q}$, we can analyze the order parameter $M_{\mathbf{Q}} = N_{\text{spin}}^{-1} \sum_{\tau} \mathbf{r}^2 \exp(-i \mathbf{Q} \cdot \mathbf{r})$ and the corresponding Binder parameter $U_{\mathbf{Q}} = \langle |M_{\mathbf{Q}}|^4 \rangle / \langle |M_{\mathbf{Q}}|^2 \rangle^2$ to determine the transition point [44]. In the ANNNI-like systems as in the model in this work, where $\mathbf{Q}$ in the thermodynamic limit can be an incommensurate wavevector, we need a modified approach since a finite-size system cannot realize any strictly incommensurate ordering.

At $T \approx T_{SDW}$, a finite-size system in such a system with periodic boundary conditions normally develops strong spin correlation at an available commensurate wavevector near the true ordering wavevector, $\mathbf{Q}_\ell(T) \approx \mathbf{Q}$. This wavevector can be detected as a peak in the spin structure factor $S_{\mathbf{q}} = N_{\text{spin}}(\langle |M_{\mathbf{q}}|^2 \rangle - \langle |M_{\mathbf{Q}}|^2 \rangle)$ and $\lim_{L \to \infty} \mathbf{Q}_\ell(T) = \mathbf{Q}(T)$ is expected. Therefore, we first evaluate $\mathbf{Q}_\ell(T)$ as the size- and $T$-dependent effective ordering wavevector, as shown in the main text, and then also $U_{\mathbf{Q}_\ell(T)} = \langle |M_{\mathbf{Q}_\ell(T)}|^4 \rangle / \langle |M_{\mathbf{Q}_\ell(T)}|^2 \rangle^2$ at each temperature $T$. The results are demonstrated in Fig. 4(a), Fig. 5(a), and Fig. 6(a) for $h/|J_1| = 0, 0.05, 0.2$, and 0.2, respectively. The modified approach allows us to identify $T_{SDW}$ as the first crossing point as lowering temperature for different system sizes [Fig. 4(a), Fig. 5(a), and Fig. 6(a)]. However, some degrees of finite-size variance in $\mathbf{Q}_\ell(T)$ may be present upon a closer investigation near the precise crossing point, as seen in Fig. 4(b), Fig. 5(b), and Fig. 6(b). For example, when $\mathbf{Q}_\ell(T)$ changes from one value to another as a function of $T$, a disordering effect can be induced at large distance, which results in a spurious peak in $U_{\mathbf{Q}_\ell(T)}$ [Fig. 4(a), Fig. 5(a), and Fig. 6(a)]. Such a spiky behavior can also affect the behavior near the crossing point, and hence, can interfere with its high-precision assessment to determine $T_{SDW}$ very accurately. However, we find that a precision achieved by treating the first crossing points of $U_{\mathbf{Q}_\ell(T)}$ for $(L_c, 2L_c) = (80, 160), (120, 240)$, and $(160, 320)$ simply on an equal footing suffices for our purpose in this work. We

**FIG. 4.** MC results for $h = 0$: (a) the Binder parameter for the order parameter with the wavevector $\mathbf{q} = \mathbf{Q}_\ell(T)$, where $\mathbf{Q}_\ell(T)$ is the size- and $T$-dependent effective ordering wavevector (see the text), (b) an enlarged view thereof near $T = T_{SDW}$, (c) the Binder ratio for the commensurate order parameter for the wavevector $\mathbf{q} = \mathbf{Q}_{\text{com}}$, (d) the square of the commensurate order parameter, and (e) the $C_6$ indicator to distinguish between the PDA state ($C_6 < 0$) and the FIM state ($C_6 > 0$).

**FIG. 5.** MC results for $h/|J_1| = 0.05$: (a) the Binder parameter for the order parameter with the wavevector $\mathbf{q} = \mathbf{Q}_\ell(T)$, where $\mathbf{Q}_\ell(T)$ is the size- and $T$-dependent effective ordering wavevector (see the text), (b) an enlarged view thereof near $T = T_{SDW}$, (c) the Binder ratio for the commensurate order parameter for the wavevector $\mathbf{q} = \mathbf{Q}_{\text{com}}$, (d) the square of the commensurate order parameter, and (e) the $C_6$ indicator to distinguish between the PDA state ($C_6 < 0$) and the FIM state ($C_6 > 0$).
transition and a direct transition. We obtain Fig. 6(c). We can apply this method for both an IC-C

obtain, e.g., $T_{\text{SDW}}/|J_1| = 1.407(5), 1.406(1),$ and $1.41(2)$ for $h/|J_1| = 0, 0.05,$ and $0.2,$ respectively.

Supplementary Material: 3. Determination of $T_{\text{IC-C}}$ or $T_c$

To analyze transitions into the three-sublattice commensurate ordered phases, we use the Binder parameter $U_{\text{com}} = \langle |M_{\text{com}}|^4 \rangle / \langle |M_{\text{com}}|^2 \rangle^2$ for the order parameter at $q = q_{\text{com}} = (0, 0, 2\pi)$, as demonstrated in Fig. 4(c), Fig. 5(c), and Fig. 6(c). We can apply this method for both an IC-C transition and a direct transition. We obtain $T_{\text{IC-C}}/|J_1| = 0.52(1), 1.15(3),$ and $1.397(3)$ for $h/|J_1| = 0, 0.05,$ and $0.2,$ respectively. These estimates are consistent with the behavior of $\langle |M_{\text{com}}|^2 \rangle$, which increases rapidly around the estimated temperatures and becomes almost size-independent at lower temperatures, as shown in Fig. 4(d), Fig. 5(d), and Fig. 6(d). The IC-C transitions studied in the present MC work exhibit features typical of a first-order transition in many cases of $h$, such as the correction in the finite-size transition temperature varying as $\sim 1/L^3$. However, these features are probably caused by discretized wavevectors in the finite size system, and it would be too early at this stage to conclude whether or not they reflect a true thermodynamic behavior. Finally, by measuring the $C_6$ indicator mentioned in the main text, we find that the ordered state for $h = 0$ is the PDA state, while we find the FIM $\uparrow\downarrow\downarrow$ state in every investigated $h \neq 0$ case down to $h/|J_1| = 0.025$.

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