Magnon dispersion and dynamic spin response in three-dimensional spin models for $\alpha$-RuCl$_3$

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In the search for experimental realizations of bond-anisotropic Kitaev interactions and resulting spin-liquid phases, the layered magnet $\alpha$-RuCl$_3$ is a prime candidate. Its modelling typically involves Heisenberg, Kitaev, and symmetric off-diagonal $\Gamma$ interactions on the two-dimensional honeycomb lattice. However, recent neutron-scattering experiments point towards a sizeable magnetic interlayer coupling. Here we study three-dimensional exchange models for $\alpha$-RuCl$_3$, for both possible $R\bar{3}$ and $C2/m$ crystal structures. We discuss the symmetry constraints on the interlayer couplings, construct minimal models, and use them to compute the magnetic mode dispersion and the dynamical spin structure factor, in both the zero-field zigzag phase and the paramagnetic high-field phase. Our predictions for the interlayer mode dispersion shall guide future experiments; they also call for a reevaluation of the quantitative model parameters relevant for $\alpha$-RuCl$_3$.

I. INTRODUCTION

Mott-insulating magnets with strong spin-orbit coupling have become a major research field in condensed-matter physics.\textsuperscript{1,2} This has been partially triggered by Kitaev’s construction\textsuperscript{3} of a quantum spin liquid driven by particular bond-anisotropic exchange interactions on the honeycomb lattice, and by the subsequent proposal\textsuperscript{4} to realize Kitaev interactions in layered honeycomb magnets with $j_{\text{eff}} = 1/2$ moments.

Among the candidate materials, $\alpha$-RuCl$_3$ has received enormous interest. It displays low-temperature antiferromagnetic order of zigzag type, and this order can be suppressed by a moderate in-plane magnetic field.\textsuperscript{5–12} By now, the existence of a quantum spin-liquid phase in $\alpha$-RuCl$_3$ in a narrow window of magnetic fields is suggested by a number of experimental results, such as an excitation continuum in neutron scattering,\textsuperscript{13,14} a transition signature in magnetocaloric-effect measurements\textsuperscript{15} and, most prominently, an approximately half-quantized thermal Hall conductivity,\textsuperscript{15,16} signifying the presence of a Majorana edge mode.

$\alpha$-RuCl$_3$ belongs to a family of layered van-der-Waals crystals, and due to the weak bonding between the layers its three-dimensional crystal structure appears to be fragile. While it adopts\textsuperscript{6,17} a monoclinic structure with space group $C2/m$ at room temperature, the low-temperature structure has been a matter of debate.\textsuperscript{2} Here, three different structures have been reported, namely monoclinic $C2/m$, trigonal $P3_112$, and rhombohedral $R\bar{3}$; they are distinguished by the pattern and sequence of the stacking of the honeycomb layers.\textsuperscript{17–20} Experimentally, stacking faults appear frequently, which also significantly influence the magnetic properties: Early samples displayed two thermodynamic transitions at $T_N = 8$ K and 14 K, while more recent higher-quality samples show a single transition at $7$ K.\textsuperscript{5,17,21} For some recent samples, a structural phase transition was found\textsuperscript{22–25} around 100–150 K, with the refinement of the neutron-diffraction data consistent with the rhombohedral $R\bar{3}$ structure at low temperature.\textsuperscript{19}

Most theoretical descriptions of the magnetism of $\alpha$-RuCl$_3$ have been restricted to planar exchange Hamiltonians, with the magnetic interlayer coupling assumed to be negligible. In contrast, recent inelastic neutron-scattering data\textsuperscript{15} indicate a significant out-of-plane dispersion of magnetic excitations. This calls for a modelling of the relevant interlayer interactions and their consequences, which is lacking to our knowledge.

It is the purpose of this paper to close this gap. For the two most probable crystal structures $R\bar{3}$ and $C2/m$, we shall discuss symmetries and corresponding minimal models for the magnetic interlayer couplings. We then employ spin-wave theory to calculate the three-dimensional magnetic mode dispersion and the dynamic spin structure factor, both in the zero-field zigzag phase as well as in the high-field phase. Comparing our results to experimental data, we obtain a consistent description of a variety of experimental data for an assumed $R\bar{3}$ crystal structure at low $T$. Our results provide concrete predictions for future experiments and will help constraining the model parameters relevant for $\alpha$-RuCl$_3$: The sizeable interlayer coupling cannot be neglected when fitting experimental data, and consequently estimates for the intralayer couplings from previous modelling need to be revised.

The remainder of the paper is organized as follows: In Sec. II, we summarize the structural models put forward for $\alpha$-RuCl$_3$ and discuss the magnetic exchange Hamiltonians, with focus on the $R\bar{3}$ and $C2/m$ structures. Section III discusses the application of spin-wave theory. In particular, we use analytical results in the high-field phase together with available experimental data to constrain the model parameters. In Sec. IV, we then display numerical results for the full mode dispersion and the dynamic structure factor and discuss them vis-à-vis published experimental data. A summary of our results, together with suggestions for future experiments, closes the paper.

II. CRYSTAL STRUCTURE AND EXCHANGE MODELS

A. Intralayer exchange Hamiltonian

The two-dimensional spin Hamiltonians proposed for $\alpha$-RuCl$_3$ are extensions\textsuperscript{26} of the honeycomb-lattice Heisenberg-Kitaev model originally introduced in Ref. 4. For the purpose of this paper, we will consider the following in-plane...
exchange interactions

\[
\mathcal{H}_0 = \sum_{n,(ij)_{\gamma}} \left[ J_0 \mathbf{S}_{n,i} \cdot \mathbf{S}_{n,j} + KS_{n,i}^{\gamma}S_{n,j}^{\gamma} + \Gamma \left( S_{n,i}^{x}S_{n,j}^{x} + S_{n,i}^{y}S_{n,j}^{y} \right) \right] + \sum_{n,(ij)_{\|}} J_3 \mathbf{S}_{n,i} \cdot \mathbf{S}_{n,j} - \vec{H} \cdot \sum_{n,i} \mathbf{S}_{n,i},
\] (1)

where \( J \) and \( J_3 \) correspond to first- and third-neighbor Heisenberg couplings, while \( K \) and \( \Gamma \) are the first-neighbor Kitaev and symmetric off-diagonal couplings, respectively. \( n \) is the layer index, and \( (ij)_{\gamma} \) denote first-neighbor \( \gamma \) bonds, with \( \gamma = x,y,z \). On \( z \) bonds \( (\alpha,\beta,\gamma) = (x,y,z) \), with cyclic permutation for \( x \) and \( y \) bonds. The uniform magnetic field is \( \vec{H} := g\mu_B\mu_0\vec{H} \), with \( g \) the (possibly anisotropic) effective g tensor and \( \mu_0 \) the Bohr magneton.

Within each layer, we assume a perfect honeycomb structure, i.e., neglect possible trigonal distortions. Then, \( \mathcal{H}_0 \) has a \( C_{3v} \) symmetry which combines a 120° real-space rotation about a site with a spin rotation about the [111] direction in spin space, exchanging \( x \rightarrow y \rightarrow z \rightarrow x \). This symmetry also implies that the experimentally relevant zigzag state is threefold degenerate, with three symmetry-equivalent in-plane propagation directions.

For the extended Heisenberg-Kitaev-\( \Gamma \) model (1), different parameter sets have been proposed to describe \( \alpha \)-RuCl\(_3\), based on either ab-initio modelling or on fits to experimental data, and we refer the reader to Ref. 28 for an overview. Guided by previous work,10,20,28–33 we employ parameters where \( K < 0 \) and \( \Gamma > 0 \) are the dominant couplings, while both \( J < 0 \) and \( J_3 > 0 \) are small, mainly acting to stabilize the zigzag phase. As will become clear below, the quantitative choice of the in-plane model parameters needs to be revisited upon including significant inter-layer interactions.

B. \( \bar{R}3 \) structure and interlayer interactions

The rhombohedral structure with \( \bar{R}3 \) space group has a conventional crystallographic unit cell consisting of three honeycomb layers. The layers are stacked with a \((2a + b - c)/3 \) translation, Fig. 1, such that the \( \bar{C}_3 \) rotation symmetry is preserved and the honeycomb lattice is undistorted. A second crystallographic domain, dubbed reverse-obverse twin,34 can be obtained by a reflection in the \( ab \) plane. From neutron diffraction,17,35 it is known that the low-field zigzag phase in the samples with a single transition at \( T_N = 7 \) K exhibits a magnetic unit cell of three layers. This implies a stacked magnetic order as shown in Fig. 2(a), which we dub 3f-zz.

We proceed by discussing a minimal model for magnetic interlayer couplings. Each spin has one interlayer neighbor which is located either right above or below it, depending on the sublattice index. This vertical spin-spin exchange interaction is compatible with the \( \bar{C}_3 \) symmetry only for a Heisenberg coupling, denoted as \( J_{\perp 1} \) in Fig. 1(a). Further, each spin has nine next-nearest interlayer neighbors which fall into two classes (with six and three members, respectively) that are distinguished by the presence or absence of a nearest-neighbor intralayer bond in one of the participating layers. In the spirit of a minimal model, we will not distinguish between these different next-nearest interlayer neighbors, and assume Heisenberg interactions, \( J_{\perp 2} \), although spin-anisotropic interactions are symmetry-allowed here. The interlayer part of the Hamiltonian thus reads

\[
\mathcal{H}_{1}^{\bar{R}3} = J_{\perp 1} \sum_{(m,n)} S_{n,j} \cdot S_{m,j} + J_{\perp 2} \sum_{(m,n)} S_{n,j} \cdot S_{m,j} \tag{2}
\]

where the number above the summation symbol indicates the number of terms per spin.

Assuming that the nearest-neighbor interlayer bonds \( J_{\perp 1} \) dominate the interlayer exchange, the 3f-zz configuration in Fig. 2(a) requires an antiferromagnetic interlayer coupling, \( J_{\perp 1} > 0 \). We note that a ferromagnetic coupling would lead to
The ordered moment is of Heisenberg type implies that the direction of the zero-field honeycomb layers. Here, the layers are stacked with a conventional trigonal unit cell that consists of three layers. According to the ab-initio analysis of Ref. 20, three types of inter-layer couplings are important and comparable in strength, namely first-neighbor bonds, such as those along (a + c)/3 and (b + c)/3, which we assume to be of equal strength, and second-neighbor bonds along (2b − c)/3. Although the form of these interactions is not symmetry-restricted, we confine ourselves to Heisenberg couplings $J_{\perp1}$ and $J_{\perp2}$, see Fig. 1(b),

$$\mathcal{H}_{1}^{C2/m} = J_{\perp1} \sum_{\langle i,j \rangle} \vec{S}_{i} \cdot \vec{S}_{j} + J_{\perp2} \sum_{\langle i,j \rangle} \vec{S}_{i} \cdot \vec{S}_{j},$$

(3)

Assuming again that the interlayer bonds obey $|J_{\perp1}| \gg |J_{\perp2}|$, realizing the 3f-zz order here requires ferromagnetic interlayer couplings, Fig. 2(b), as opposed to the antiferromagnetic couplings necessary in the $R3$ structure.

Two remarks are in order: (i) Since the $C2/m$ structure breaks the $C_3$ symmetry, the three propagation directions of the zigzag order are in general no longer degenerate. They remain, however, degenerate at the classical level within our model if we set $J_{\perp2} = 0$. (ii) If instead the interlayer coupling $J_{\perp2}$ dominates over $J_{\perp1}$, then an antiferromagnetic $J_{\perp2}$ may induce, depending on the sign of $J_{\perp1}$, either a 6f-zz state propagating perpendicular to the $z$-bond or a 3f-zz state propagating perpendicular to the $x$ or $y$ bond. We will not explore this option in detail.

III. SPIN-WAVE THEORY AND PARAMETER CHOICE

For the models of Sec. II, we employ standard linear spin-wave theory for spins of size $S$ on the two different lattices, see the Supplemental Materials to Refs. 10 and 37 for details. We calculate the dynamic spin structure factor at $T = 0$ according to

$$S(q, \omega) = \sum_{\alpha} \int d\tau e^{i\omega \tau} \langle S^{\alpha}(q, \tau)S^{\alpha}(-q, 0) \rangle.$$  

(4)

When specifying momenta, we will follow the conventions of Refs. 13 and 14 and use reciprocal-space coordinates ($H, K, L$) in reciprocal lattice units (rlu), corresponding to an embedding trigonal unit cell. In this convention, the in-plane $M$ point is located at $(H, K) = (0, 1/2)$ while the in-plane $K$ point is at $(-1/3, 2/3)$. For the vertical direction, this convention implies that mode energies will be $L$-periodic with a period of 3 in the 3f-zz magnetic structure.

For discussing the parameter dependence of the results and relate them to experimental data, it is useful to define an overall energy scale $A$ and parameterize the couplings as $(J, K, \Gamma, J_{\perp1}, J_{\perp2}) = A(J, K, \Gamma, J_{\perp1}, J_{\perp2})$. Similarly, we define the strength of the magnetic field as $|\vec{h}| = AS\hat{h}$.

A. Phases

In the high-field phase, the spin-wave expansion is performed about the polarized state. We will show results for magnetic fields along the two crystallographic in-plane directions perpendicular and parallel to a Ru-Ru bond, which correspond to the $(1, -2, 0)$ and $(1, 0, 0)$ directions in the reciprocal-space basis, respectively. In both cases, the magnetization in the high-field phase points along the field direction even in the presence of a finite $\Gamma$ term. We work with a two-site unit cell, and the linear-spin-wave calculation amounts to performing a $4 \times 4$ Bogoliubov transformation, for details see Ref. 39.

In the zigzag phase, we work exclusively at zero field. The spin-wave expansion is performed about a 3f-zz state, where the direction of the ordered moment is fixed by the ratio of $\Gamma$ and $K$, see Sec. VI of Ref. 28 for details. In particular, for $\Gamma/K = 1/2$ as used below, the magnetic moments $g\vec{S}$ point at an angle of $25^\circ$ out of plane if a $g$-factor anisotropy of $g_{ab}/g_c = 1.77$ is used. The magnetic unit cell contains four sites and is periodically repeated in each layer, such that an $8 \times 8$ Bogoliubov transformation needs to be performed. As
noted above, for the $R^3$ structure, there are three symmetry-equivalent propagation directions of the zigzag order. We perform the calculation of the spin structure factor for one of the three zigzag domains, obtain the result for the other domains by a $C_3$ rotation, and average the result over all three possible domains. For the $C2/m$ structure, this equivalence is violated, and we consider only the energetically favorable zigzag state with propagation direction perpendicular to the $z$ bond (assuming ferromagnetic interlayer couplings), without domain averaging.

### B. Parameter choice

Before discussing in detail numerical data obtained for concrete parameter sets, Figs. 3-5, we find it useful to derive a few analytical results. In fact, for the high-field phase, one can determine the maxima and minima of the interlayer dispersion of the lowest magnon mode in closed forms. We will use these together with the experimental information from Ref. 14 to derive constraints on the model parameters.

1. **R3 structure**

For the $R^3$ structure described by the model in Eq. (2), we assume $J_{1,2} > 0$, which yields 3f-zz order. For $q = (0,0,L)$ and $\vec{h} \parallel (1,-2,0)$, the energy of the lowest mode in the high-field phase can be calculated as function of $L$. It takes its extremal values at $L = 0$ and $L = 1.5$ in rlu. For $J_{1,2} > 0$, the maximum is at $L = 0$ and reads

$$\omega_{\text{max}}^2/(AS)^2 = \hat{h}(\hat{h} + 3\hat{\Gamma})$$

which, remarkably, does not depend on the interlayer coupling. In the high-field limit, $\omega_{\text{max}} \rightarrow |\hat{h}|$ as expected. Similarly, the minimum energy $\omega_{\text{min}}$ is taken at $L = 1.5$ and evaluates to

$$\omega_{\text{min}}^2/(AS)^2 = (\hat{h} - 2J_{1,1} - 18J_{1,2})(\hat{h} - 2J_{1,1} - 18J_{1,2} + 3\hat{\Gamma}).$$

We can also compute the critical field for the disappearance of the zigzag order; it is given by

$$\hat{h}_c = 2\hat{J} + \hat{K} - \frac{\hat{\Gamma}}{2} + 6\hat{\Gamma} + 10\hat{J}_{1,2} + \sqrt{\hat{K}^2 - \hat{\Gamma}^2 + \frac{9}{4}}$$

for $\hat{h} \parallel (1,0,0)$; this is the direction for which the additional ordered phase found in Ref. 33 is either absent or very narrow. Eq. (7) underlines the collective role played by $J_3$, $J_{1,1}$, and $J_{1,2}$ in stabilizing the zigzag order, as all of them contribute to increase the critical field.

The experiment of Ref. 14 has determined the mode dispersion by inelastic neutron scattering, with the results $\omega_{\text{max}} \approx 4.3$ meV and $\omega_{\text{min}} \approx 3$ meV for $B = 13.5$ T applied along the $(1,-2,0)$ direction. Moreover, the experimental critical field is about $B_c = 7.6$ T for the $(1,0,0)$ direction. Together, this information can be used to constrain the model parameters; further constraints arise from fitting the zero-field interlayer dispersion, see below. We note that accurate information on the in-plane spin-wave dispersion in $\alpha$-RuCl$_3$ is available neither at zero field nor at high fields, leaving a considerable uncertainty in a conclusive determination of model parameters from mode dispersions.

To build plausible sets of model parameters, we first fix the ratio of the in-plane nearest-neighbor couplings to $\hat{J} : \hat{K} : \hat{\Gamma} = -0.1 : -1 : 0.5$, as obtained in Ref. 30. We then choose ratios of $J_3/\hat{K}$ and $J_{1,2}/J_{1,1}$. With these fixed, $A$ and $J_{1,1}$ are uniquely determined by matching $\omega_{\text{min}}$ and $\omega_{\text{max}}$ at $\hat{h}/\hat{h}_c = 1.78$ with the experimental data at 13.5 T. Finally, the value of $g_{ab}$ is determined by demanding that the critical field matches the experimental value, $B_c = 7.6$ T. Choosing $J_3/\hat{K}$ and $J_{1,2}/J_{1,1}$ is not unique: To guide this, we monitor the numerical result at zero field (where we have no analytic solution) and try to match the experimental mode dispersion with the corresponding results of Ref. 14. We also observe that the quality of this match depends only weakly on $J_{1,2}/J_{1,1}$.

Out of the family of possible parameter sets, we present results for two, which we dub Model 1 and Model 2, with the numerical parameter values shown in Table I. In both cases, the in-plane parameters deviate substantially from the ones used before. Either $J_3$ is significantly smaller (Model 2), or all parameters are significantly smaller at the expense of a larger $g$ factor (Model 1). This mainly reflects the fact that the interlayer coupling, the sizable magnitude of which is dictated by the experimentally detected out-of-plane magnon dispersion, tends to stabilize the zigzag order and hence increases the critical field.

| # | Strct | $J_3$ | $K$ | $\Gamma$ | $J_{1,1}$ | $J_{1,2}$ | $A$ [meV] | $g_{ab}$ | $h_c$ |
|---|---|---|---|---|---|---|---|---|---|
| 1 | $R^3$ | $-0.1$ | $-1$ | $0.5$ | $0.1$ | $0.3$ | $0.015$ | $2.8$ | $4.3$ | $1.34$ |
| 2 | $R^3$ | $-0.1$ | $-1$ | $0.5$ | $0.01$ | $0.08$ | $0.001$ | $10$ | $2.5$ | $0.22$ |
| 3 | $C2/m$ | $-0.1$ | $-1$ | $0.5$ | $0.1$ | $-0.05$ | $-0.05$ | $4$ | $2.7$ | $0.59$ |

2. **C2/m structure**

For the $C2/m$ structure described by the model in Eq. (3), we now assume $J_{1,2} < 0$ in order to obtain 3f-zz order. The extremal energies of the lowest mode dispersion for $q = (0,0,L)$ and $\vec{h} \parallel (1,-2,0)$ in the high-field phase are again taken at $L = 0$ and $L = 1.5$. However, for $J_{1,2} < 0$, the point $L = 0$ now represent the dispersion minimum, with

$$\omega_{\text{min}}^2/(AS)^2 = (\hat{h} + 2J_{1,2})(\hat{h} + 2J_{1,2} + 3\hat{\Gamma}).$$

Conversely, the maximum energy is now taken at $L = 1.5$ and evaluates to

$$\omega_{\text{max}}^2/(AS)^2 = (\hat{h} - 8J_{1,1} - 2J_{1,2})(\hat{h} - 8J_{1,1} - 2J_{1,2} + 3\hat{\Gamma}).$$
The critical field for the disappearance of the zigzag order is here given by

\[ \hat{h}_c = 2J + K - \frac{\Gamma}{2} + 6J_3 + \sqrt{K^2 - R\Gamma + \frac{9}{4}\Gamma} \]  

for \( \hat{h} \parallel (1, 0, 0) \). Interestingly, this is independent of the interlayer coupling as in both the canted zigzag and high-field phases all spins coupled by \( J_{\perp,2} \) are aligned in parallel fashion.

Following a matching procedure similar to the one described above, we arrive at a parameter set which we dub Model 3, see Table I. Here, the in-plane parameters are not very different from previous modelling, \(^3^8\) because the interlayer coupling is ferromagnetic and does not influence the critical field.

\[ \text{IV. NUMERICAL RESULTS} \]

We now turn to a discussion of the full numerical results for the dynamic spin structure factor, shown in Figs. 3, 4, and 5 for Models 1, 2, and 3, respectively. For comparison, we have added experimentally determined magnetic mode energies extracted from THz spectroscopy, \(^4^2\) electron spin resonance (ESR), \(^4^3\) inelastic neutron scattering (INS), \(^1^4\) and thermal transport \(^1^2\) measurements.

For both Models 1 and 2, we obtain agreement with the high-field dispersion along \((0, 0, L)\) as measured by INS; given the momentum-space location of minimum and maximum this
agreement is achieved by construction. Model 1 also displays excellent agreement for the zero-field dispersion along (0, 0, L) as well as with the THz and ESR data; Model 2 performs inferior in this respect. We note that Model 1 has a rather small overall energy scale $\Delta$ and a large $g$ factor.

In contrast, Model 3 fails to match the high-field dispersion along (0, 0, L). This is because the momentum-space location of minimum and maximum are switched w.r.t. Models 1 and 2, because the interlayer coupling is ferromagnetic here, $J_{1,2} < 0$. We recall that this is required in order to stabilize the experimentally observed $3f$-zz magnetic order.

Given the mismatch visible in Fig. 5, we conclude that a $C2/m$ low-temperature crystal structure appears unlikely to be realized in $\alpha$-RuCl$_3$. Instead, the assumption of an $R3$ crystal structure with sizeable antiferromagnetic interlayer couplings leads to results consistent with experiment.

V. SUMMARY

Motivated by recent neutron-scattering results indicating a significant interlayer dispersion, we have discussed three-dimensional spin models for the Kitaev material $\alpha$-RuCl$_3$. We have considered two candidate crystal structures, $R3$ and $C2/m$. For both, we have constructed minimal interlayer coupling models and determined the mode dispersion and dynamical spin structure factor using spin-wave theory, both in zero field and at high fields.

Our results show that the minimal models for the $C2/m$ structure cannot simultaneously reproduce the experimentally found zero-field magnetic structure and the form of the high-field interlayer dispersion. In contrast, the minimal models for $R3$ can reproduce both, provided that the interlayer couplings are assumed to be sizeable and antiferromagnetic. In fact, in both Models 1 and 2 the interlayer coupling is of order 1 meV, as dictated by the experimentally observed magnon bandwidth of 1.3 meV.

FIG. 4. Same as Fig. 3, but now for Model 2 with $R3$ crystal structure. While this parameter set also reproduces the high-field mode energies (see text for details), it does not match quantitatively at zero field.
FIG. 5. Same as Fig. 3, but now for Model 3, which has C2/m crystal structure. Here, panels (a-c) have been calculated for the energetically favorable zigzag direction and do not involve domain averaging. This model cannot reproduce the qualitative shape of the high-field dispersion; this generically applies to C2/m models, which require a ferromagnetic interlayer coupling in order to stabilize threefold-periodic zigzag order in zero field.

For simplicity, we have neglected spin anisotropies in the interlayer interactions. While those will be present (except for the vertical coupling in the R3 structure) and will change the detailed quantitative fitting of experimental data, we expect our analysis to be semi-quantitatively robust concerning the magnitude of the couplings. Interlayer interaction anisotropies are likely to play a role for the intermediate ordered phase observed in α-RuCl3; this will be explored in a forthcoming publication.36

Our analysis clearly shows that interlayer interactions cannot be neglected in α-RuCl3 when it comes to quantitative modelling, because these interactions substantially influence the stability of the zigzag phase. We also note that sharp magnon modes will receive a broadening of the order of the interlayer magnon bandwidth, if scattering data are integrated over substantial ranges of the out-of-plane momentum. More detailed neutron-scattering studies are therefore called for. In particular, the interlayer dispersion at high fields should be measured at various in-plane wavevectors, and ideally also various field directions, which will enable to better discriminate between the parameter sets of Models 1 and 2.

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