Magnetic order in the quantum spin chain compound SrCo$_2$V$_2$O$_8$: a single-crystal neutron diffraction study in magnetic field

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We explore the spin states in the quantum spin chain compound SrCo$_2$V$_2$O$_8$ up to 14.9 T and down to 50 mK, using single-crystal neutron diffraction. Upon cooling in zero-field, antiferromagnetic (AFM) order of Néel type develops at $T_N \approx 5.0$ K. Applying an external magnetic field ($H \parallel c$-axis) destabilizes the Néel order, leading to an order-disorder transition when applying a field between $T_N$ and $\sim 1.5$ K. Below 1.5 K, we observe a Néel to longitudinal spin density wave (LSDW) order transition at 3.9 T, and a LSDW to emergent AFM order transition at 7.0 T. Our results also reveal several unique signatures for the states of the spins that are not present in the isostructural counterpart BaCo$_2$V$_2$O$_8$.

I. INTRODUCTION

Magnetic field is a very important parameter in tuning the physical properties in quasi one-dimensional (1D) spin-1/2 magnets. The magnetic excitation spectrum of a single quantum chain is a continuum composed of pairs of spinons with $S = 1/2$ that can propagate like domain walls. In quasi 1D magnets where there exist interactions between the chains, the spinons become confined by an attractive potential. Concomitantly, the continuum spectrum is replaced by a series of discrete spinon bound states. It has been found that the nature of the spinon confinement can be significantly modified by applying a magnetic field.

Exotic magnetic long-range order (LRO) may occur in a magnetic field. For example, in weakly coupled spin chains or ladders with a singlet-dimer ground state ($S = 0$), applying a magnetic field splits the triplet excitation ($S = 0, \pm 1$); a singlet-dimer to LRO transition occurs at the closure of the energy gap corresponding to the lowest triplet branch ($S = 1$). This transition, also known as magnon Bose-Einstein condensation (BEC), has been intensively studied in the last two decades.

Recently, another magnetic-field-driven quasiparticle condensation process, illustrated as the onset of the longitudinal spin density wave (LSDW) order, has been identified in BaCo$_2$V$_2$O$_8$. This compound crystallizes in a body-centered tetragonal lattice (space group $I4_1/acd$), in which screw chains of CoO$_6$-octahedra run along the crystallographic $c$-axis. These chains are well separated by Ba and Sr in the $ab$-plane, greatly reducing the strength of the interactions between the chains. The Co$^{2+}$ ion ($3d^7$) has an effective spin of 1/2 because of the octahedral distortion and spin-orbit coupling. Previous investigations have shown that the intrachain spin interactions in BaCo$_2$V$_2$O$_8$ can be well described by a XXZ model written as

$$\mathcal{H} = J \sum_i \{S_i^x S_{i+1}^x + \epsilon(S_i^y S_{i+1}^y + S_i^z S_{i+1}^z)\} - g_z \mu_B \sum_i S_i^z H,$$

where $J > 0$ is the nearest-neighbour (NN) antiferromagnetic (AFM) exchange constant, $\epsilon$ is the anisotropy parameter, and $g_z$ is the component of the Landé $g$-tensor along the chain direction. This Hamiltonian can be exactly solved by the Bethe ansatz. Notably, the $\epsilon < 1$, $\epsilon = 1$, and $\epsilon > 1$ correspond to the Ising-like, Heisenberg, and XY-like spin chains, respectively. In the Ising-like region, the magnetic ground state is AFM order of Néel type. A spin disordered state, described as a Tomonaga-Luttinger liquid (TLL), can be induced above a critical magnetic field, as this increases the 1-D character of the system. The spinon spectrum in the TLL state of an Ising-like XXZ spin chain is dominated by a longitudinal (transverse) mode in the intermediate (high) magnetic field region. The longitudinal mode is characterized by an incommensurate modulation vector ($\delta l$), which can be expressed as

$$|\delta l| = 4 \times <S_z> = 4 \times M_z / g_z \mu_B.$$

where $M_z$ is the magnetization along the chain direction. In BaCo$_2$V$_2$O$_8$, interactions between the spin chains are negligibl[e, leading to the occurrence of Néel order at a finite temperature $T_N = 5.5(1)$ K. Notably, the Néel order is unstable against an external magnetic field; a field-induced Néel to LSDW order transition occurs below $\sim 1.8$ K. Since the incommensurate modulation vector of the LSDW order strictly follows the theoretical prediction of Eq. 2, this novel state of spins has been suggested to originate from the condensation of the longitudinal spin fluctuation in a TLL.
However, their theory predicts robust LSDW order that does not collapse until 15.1 T, above which transverse staggered AFM order overwhelms the former. In sharp contrast, single-crystal neutron diffraction measurements see an LSDW to transverse AFM order transition at 8.5 T; the LSDW order completely vanishes at 9.25 T. These values significantly deviate from the theoretical prediction. This discrepancy might indicate that the inter- and intra-chain exchange parameters used in Ref. 23 are not correct or that the mean-field theory is inadequate. As pointed out by a recent nuclear magnetic resonance (NMR) study, the intra- and inter-couplings in BaCo$_2$V$_2$O$_8$ are rather complicated and could be strongly field-dependent.

To the best of our knowledge, BaCo$_2$V$_2$O$_8$ is the only known TLL system in which the spins are driven into a LSDW or transverse AFM state at low temperatures by an external magnetic field. To obtain more insights into these emergent phases, and more importantly, understand the relevant interactions, new materials are in high demand. Here, we report the ground and magnetic-field-induced states of the spins in the isostructural compound SrCo$_2$V$_2$O$_8$ up to 14.9 T ($H \parallel c$-axis) and down to 50 mK, using single-crystal neutron diffraction. Our experimental results are organized into four sections, which focus on the Néel order (Section III), field-induced order-disorder transition (Section IV), LSDW order (Section V) and emergent AFM order (Section VI), respectively. In Section VII we discuss several unique signatures of the spin states in SrCo$_2$V$_2$O$_8$ that are not present in BaCo$_2$V$_2$O$_8$ to evidence the complex physics in this compound.

II. EXPERIMENTAL METHODS

Two high quality SrCo$_2$V$_2$O$_8$ single-crystals (~3 × 3 × 6 mm$^3$) were measured in this investigation. They were grown by the spontaneous nucleation method described in Ref. 26. All single-crystal neutron diffraction measurements were carried out at the Swiss Spallation Neutron Source (SINQ) at the Paul Scherrer Institute. Both single-crystals were aligned using the two-axis neutron diffractometer ORION. The first single-crystal was mounted into a dilution refrigerator and then into a 15 T vertical cryomagnet equipped with a dilution refrigerator insert and then measured on the cold triple-axis spectrometer RITA-II. The $c$-axis was aligned along the magnetic field. The incident neutron energy was fixed at 5 meV using a vertically focusing PG (002) monochromator. The energy of the scattered neutrons was analysed using a multiblade PG (002) crystal analyzer, which was operated in monochromatic imaging mode. A cooled beryllium filter was placed between the sample and analyzers to suppress $\lambda/2$ contamination. The neutrons were detected using a position-sensitive detector (PSD) consisting of 128 × 128 pixels. In all measurements, only the elastic scattering signal was recorded.

III. NÉEL ORDER AT $\mu_0 H = 0$ T

A crystallographically forbidden reflection (2, 3, 0) is observed at $T = 50$ mK and $\mu_0 H = 0$ T (Fig. 1a). This corresponds to a Néel state in which the spins are modulated by $k = (0, 0, 1)$ (Fig. 1b). Fig. 1 shows cuts through this reflection, measured on TriCS. Each peak was fitted using a Gaussian function, giving full width at half maximum (FWHM) values of 0.0310(5), 0.0242(4), and 0.0981(1) r.l.u. for the $h$-, $k$-, and $l$- scans, respectively. These values serve as the TriCS instrumental resolution parameters around this momentum transfer (referred to

![Image](https://example.com/image.png)
TABLE I. Basis functions $\Psi_n$ ($n = 1, ..., 12$) of Co for the irreducible representation $\Gamma_5$. The used basis functions in the refinement are highlighted in grey. The atomic sites are labelled as Co1: $(x, y, z)$, Co2: $(-x + 1, -y + 1, z)$, Co3: $(-y + 1, x + 1/2, z + 1/4)$, Co4: $(-x + 1, y, z + 1/2)$, Co5: $(-x + 1, y, z + 1/2)$, Co6: $(x, -y + 1, z + 1/4)$, Co7: $(y + 1/2, z, z + 1/4)$, and Co8: $(-y + 1/2, x + 1, z + 1/4)$.

| Group I | $\Psi_1$ | $\Psi_2$ | $\Psi_3$ | $\Psi_4$ | $\Psi_5$ | $\Psi_6$ | $\Psi_7$ | $\Psi_8$ | $\Psi_9$ | $\Psi_{10}$ | $\Psi_{11}$ | $\Psi_{12}$ |
|---------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| Co1     | 1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   |
| Co2     | 1 0 0   | 0 1 0   | 0 0 -1  | 1 0 0   | 0 1 0   | 0 0 -1  | 1 0 0   | 0 1 0   | 0 0 -1  | 1 0 0   | 0 1 0   | 0 0 -1  |
| Co3     | 0 1 0   | -1 0 0   | 0 0 1   | 0 -1 0  | 1 0 0   | 0 0 1   | 0 -1 0  | 1 0 0   | 0 0 1   | 0 -1 0  | 1 0 0   | 0 0 1   |
| Co4     | 0 1 0   | -1 0 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  |

| Group II | $\Psi_1$ | $\Psi_2$ | $\Psi_3$ | $\Psi_4$ | $\Psi_5$ | $\Psi_6$ | $\Psi_7$ | $\Psi_8$ | $\Psi_9$ | $\Psi_{10}$ | $\Psi_{11}$ | $\Psi_{12}$ |
|---------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| Co5     | -1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   | 1 0 0   | 0 1 0   | 0 0 1   |
| Co6     | -1 0 0   | 0 1 0   | 0 0 -1  | -1 0 0  | 0 1 0   | 0 0 -1  | -1 0 0  | 0 1 0   | 0 0 -1  | -1 0 0  | 0 1 0   | 0 0 -1  |
| Co7     | 0 1 0   | 1 0 0   | 0 0 1   | 0 -1 0  | -1 0 0  | 0 0 1   | 0 -1 0  | -1 0 0  | 0 0 1   | 0 -1 0  | -1 0 0  | 0 0 1   |
| Co8     | 0 1 0   | 1 0 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  | 0 -1 0  | 0 1 0   | 0 0 -1  |

below).

We have studied the $(2, 3, 0)$ reflection as a function of temperature ($T$). The magnetic order parameter is proportional to the square root of the integrated intensity ($I$) of the corresponding magnetic Bragg peak. So the critical exponent $\beta$, which reveals the nature of the magnetic order, obeys $I (T < T_N) = I_0 + A (1 - T/T_N)^{2\beta}$

Since the position and width of the reflection belonging to the Néel order do not change with temperature, we only measured the neutron counts on the peak position of the $(2, 3, 0)$ reflection. We have fitted the temperature dependence of the peak intensity using the above equation in a temperature range between $0.84T_N$ and $T_N$ (Fig. 1b), which produces $\beta = 0.3154(95)$ and $T_N = 5.00(5)$ K. The fitted $\beta$ is in agreement with the theoretical description for a three-dimensional Ising model in which $\beta \approx 0.326$.

To study the magnetic structure in the Néel phase, we first carried out a representational analysis using the SARAh Representational Analysis software. Five irreducible representations ($\Gamma_n$, $n = 1, 2, 3, 4, 5$) could be obtained, but $\Gamma_5$ correctly produces the Néel order in SrCo$_2$V$_2$O$_8$. In the Néel phase, the symmetry is no longer invariant under the $4_1$ screw operation, breaking the Co-sites into two independent groups (Co1, Co3, Co5, Co7, and Co8). For each group, six basis functions, $\Psi_n$ ($n = 1, ..., 12$), for the irreducible representation $\Gamma_5$ are required to fully describe the magnetic structure (Table I), meaning that 12 parameters need to be refined. However, our critical exponent analysis supports the dominant Ising anisotropy. We therefore only refine the longitudinal ($c$-component) moment on each Co-site. This criterion leaves us with $\Psi_3$, $\Psi_6$, $\Psi_9$, and $\Psi_{12}$. We have collected 17 nuclear and 82 magnetic reflections for the magnetic structure determination at 50 mK. Due to the 4-fold symmetry in the $ab$-plane, two magnetic domains are allowed; they are illustrated in Fig. 2. We constrain the two groups of Co-sites, e.g. Co3 and Co8 in Fig. 2b, to have an identical amplitude of magnetic moment, while their orientations are decided by symmetry. Only the scale factor, domain fractions and amplitude of the magnetic moment along the $c$-axis were refined; the atomic positions and isotropic atomic displacement parameters were fixed to the values at 1.5 K reported by Bera et al.

We then performed the refinements using the FullProf package. Initially, we included all four basis functions in our refinement. However, we could not detect any contribution from $\Psi_5$ and $\Psi_{12}$ within the resolution. This is consistent with the Néel state of Eq. 1 wherein the spins are antiferromagnetically coupled along the chain; $\Psi_5$ and $\Psi_{12}$ do not fit this description, thus, only the eligible $\Psi_3$ and $\Psi_9$ were adopted in our final refinement. The quality of our refinement is shown in Figure 2b. The data supports the critical exponent $\beta$ = 0.3154(95) and $T_N = 5.00(5)$ K.
main populations in our sample are 42(3)% and 58(3)% for Domain # 1 and Domain # 2, respectively. The refined longitudinal magnetic moment is $1.81(4) \mu_B$ per Co. We note that this value is lower than the $2.1-2.3 \mu_B$ per Co at 1.5K reported in Ref. 28, in which the same model is applied to refine the neutron powder diffraction pattern.

We tested a single-domain solution in which the aforementioned moment constraint between the two groups of Co-sites (Table I) is released. We obtained 2.6(1) and -0.20(9) $\mu_B$ per Co for the groups containing Co$_3$ and Co$_8$ (Table I), respectively. However, this single-domain scenario is not supported by the NMR study of Kawasaki et al. 33

IV. MAGNETIC FIELD INDUCED ORDER-DISORDER TRANSITION

Magnetization and heat capacity measurements in a magnetic field $H \parallel c$-axis suggest that SrCo$_2$V$_2$O$_8$ undergoes a field-induced order-disorder transition between $T_N$ and 2.0K. We studied the magnetic field dependence of the (2, 3, 0) reflection using the PSD on RITA-II. A typical diffraction pattern measured at 4.0K and 0.5T is shown in Fig.3a, in which the white frames mark the effective detector area covered by the three central analyser blades. The streak-like feature of the diffraction pattern comes from the artifact of the vertically focusing monochromator. To quantitatively analyse the data, we summed the neutron counts captured within each vertical array of detectors. The summed intensity as a function of horizontal pixel number is plotted in Fig.3b for $T = 4.0K$. Only the central frame captures the Bragg diffraction. Figure 3 plots the magnetic field dependence of the summed intensity of the (2, 3, 0) reflection, obtained by summing the normalized neutron counts captured in the central frame (Fig. 3a), at different temperatures. A field-induced order-disorder transition is clearly revealed.

We could not detect any field-induced LSDW order at the temperatures discussed in this section.

V. MAGNETIC FIELD INDUCED LONGITUDINAL SPIN DENSITY WAVE ORDER

We now study the magnetic field response of the Néel phase at temperatures down to 50mK. Figure 4 shows the (2, 3, $l$) reflection at 75mK measured between 3.2T and 5.95T on TricSl. Below 3.9T, this reflection hardly changes and is centered at $l = 0$ (Fig. 4a). The average FWHM of the peaks in this region is 0.098 r.l.u.; it is equal to the instrumental resolution within the errors (see Section III). At 3.9T, the intensity weakens (Fig. 4a). At higher fields, we see two peaks at finite $l$ values, as expected for the LSDW order.

For the 3.9T data, we have fitted three models. In the first model, we apply a single Gaussian function centered at $l = 0$ to this profile, meaning that the system is still in the Néel state. We obtain a broadened reflection with FWHM = 0.105(2) r.l.u. (Fig. 4b). In the second model, we apply two Gaussian functions centered at $l = \pm \delta l$. This model, which produces a FWHM of 0.09(1) r.l.u., corresponds to a single LSDW phase (Fig. 4b). The third model, in which we assume the coexistence of the Néel and LSDW peaks that share the same FWHM, gives a FWHM of 0.07(1) r.l.u.; we therefore rule out the third model. While the second model fits the observations in BaCo$_2$V$_2$O$_8$, 7,8 we cannot rule out the presence of a short-range ordered Néel phase at 3.9T (the first model).

We could only resolve the LSDW phase clearly when $\mu_0 H \geq 4.0T$, based on which we conclude that the LSDW state sets in around 3.9T in SrCo$_2$V$_2$O$_8$. The incommensurate peak is resolution limited in a $l$-scan at all magnetic fields measured, indicating long-range spin correlation along the $c$-axis. This is consistent with the
As the magnetic field increases, the two incommensurate peaks move apart, as expected (Fig. 4c). It has been proposed that the LSDW order in BaCo$_2$V$_2$O$_8$ results from the condensation of the longitudinal spin fluctuations in the TLL state. In BaCo$_2$V$_2$O$_8$, the incommensurate modulation vector (δl) follows Eq. 2. To check this scenario in SrCo$_2$V$_2$O$_8$, we compare our fitted δl at 75 mK with the values predicted by Eq. 2 (Fig. 5a). We used g = 6.1 along the c-axis, the Van Vleck paramagnetism correction factor $0.014 \mu_B/T$, and the longitudinal magnetization $M_z$, as reported in Ref. 35, to produce the theoretical δl. In sharp contrast to the good match between the experiment and theory in BaCo$_2$V$_2$O$_8$, all the observed δl values in SrCo$_2$V$_2$O$_8$ are much lower than the predicted values (Fig. 5a). This discrepancy will be discussed in Section VII.

We also investigated the transverse spin correlation. Unlike the three-dimensional long-range LSDW order in BaCo$_2$V$_2$O$_8$, the (2, 3, ±δl) peak is not resolution limited along both the h- and k-directions in SrCo$_2$V$_2$O$_8$ (Fig. 5b & c measured on TriCS and Fig. 8a measured on RITA-II), which suggests short-range spin correlation in the ab-plane. All these features indicate that the LSDW order in SrCo$_2$V$_2$O$_8$ is distinct from that in BaCo$_2$V$_2$O$_8$.

VI. MAGNETIC FIELD INDUCED EMERGENT ANTIFERROMAGNETIC ORDER

To explore the evolution of the LSDW order above 5.95 T in SrCo$_2$V$_2$O$_8$, we performed our measurements on RITA-II using a 15 T vertical cryomagnet. Neither the cryomagnet nor detector could be tilted, meaning that the neutron scattering intensity from the LSDW order will be weakened by the Lorentz factor. However, by taking advantage of the PSD, we were still able to resolve a partial LSDW diffraction spot, and thus track its evolution.

For the detector area enclosed by the central frame in Fig. 5a, we first summed the neutron counts along the horizontal pixel direction. The summed intensity versus
The onset of transverse AFM order in SrCo$_2$O$_8$ (Fig. 6 and Fig. 7a). This phenomenon is consistent with much weaker (2, 3, 0) reflection is recovered above 7.0T in the LSDW region (Fig. 4 and Fig. 6a). Interestingly, a δl pixel number for the (2, 3, 0) reflection as measured at 120 mK. (a) Raw data recorded at $\mu_0 H = 2.5$, 5.0 and 7.0 T, respectively. The grey box marks the effective detector coverage along the vertical direction. The green shaded area marks the additional intensity seen in the 7.0 T pattern. (b) Intensity versus vertical pixel number at $\mu_0 H = 2.5$ and 14.85 T after subtracting the 7.0 T background pattern and performing 3-pixel binning on the raw data. A scale factor of 10 has been applied to the 14.85 T curve. Inset: Intensity versus horizontal pixel number curve at 7.0 T. The grey boxes mark the effective detector coverage along the horizontal direction.

Vertical pixel number curves are plotted in Fig. 3 for T = 120 mK. As shown in Fig. 3a, the (2, 3, 0) peak is clearly split at 5.0 T, corresponding to the LSDW order. The peak on the right hand side of the commensurate position could always be resolved below 7.0 T. Unfortunately, the left hand peak goes out of the effective detector area along the vertical direction above 5.0 T. The LSDW order could be detected up to 6.5 T. For the 7.0 T pattern obtained in this particular set of measurements, we compared the neutron counts in the central frame with those obtained in the two nearest neighbouring frames (only background is present in these frames, see Fig. 3a & b) along the horizontal pixel direction (inset of Fig. 3b), we conclude that they come from the background within our resolution. Note that additional neutron counts are recorded at 7.0 T between pixel number 80 and 92 compared to the pattern obtained at 2.5 T (green shaded area of Fig. 3b). However, the 7.0 T diffraction profile is rather flat in the whole central frame. Moreover, the counting time in the LSDW region is 100 times longer than that in the Neél region. It is therefore expected that more background counts would be captured. We therefore conclude that these additional neutron counts are not of magnetic origin.

In the Neél state, the (2, 3, 0) pattern is always centered between 60 and 64 along the vertical pixel direction (Fig. 3a). This reflection splits by δl along the c*-axis in the LSDW region (Fig. 4 and Fig. 5). Interestingly, a much weaker (2, 3, 0) reflection is recovered above 7.0 T (Fig. 5a and Fig. 5b). This phenomenon is consistent with the onset of transverse AFM order in SrCo$_2$V$_2$O$_8$ at high fields.  

We plot the intensity of the detector area enclosed by the central white frame (see Fig. 3a) versus magnetic field curve in Fig. 7a. The drastic drop in intensity when $\mu_0 H \geq 4.0$ T fits the Neél to LSDW order transition discussed in Section V. We note that the rate of this drop is overestimated due to the fact that only the partial double peak profiles could be resolved above 5.0 T (Fig. 5b). The intensity reaches its minimum at 7.0 T, after which it increases monotonously with the magnetic field until 12.5 T. This behaviour also agrees well with the feature of the transverse AFM order observed in BaCo$_2$V$_2$O$_8$.  

This emergent AFM order in SrCo$_2$V$_2$O$_8$ is fully stabilized at 12.5 T and above; the intensity remains unchanged up to the highest field probed (14.9 T, see Fig. 7b).

As mentioned in Section V, the spins in the LSDW state of SrCo$_2$V$_2$O$_8$ are short-range correlated in the ab-plane (Fig. 5a & c). To further verify this effect, we repeated these measurements on RITA-II at 120 mK. The field dependences of h- and k-scans reveal the same feature. For clarity, we only display the FWHM obtained from the h-scans in Figure 6. It is clear that the (2, 3, 0) reflection is broadened along the h- and k- directions in the LSDW region, in excellent agreement with our observations on TriCS. The long-range spin correlation is recovered along both a- and b- axes when the emergent AFM order is induced.

VII. DISCUSSION

A longitudinal mode of spins, which exists as a sequence of discrete dispersing lines, is the component of the spinon spectrum in SrCo$_2$V$_2$O$_8$ and BaCo$_2$V$_2$O$_8$. This mode causes fluctuation in the amplitude of the magnetic moment, hence weakening the statically ordered value. In Section III we report that Sr- and Ba-Co$_2$V$_2$O$_8$ shares the same magnetic structure in the Neél state. However, the ordered magnetic moment per Co in SrCo$_2$V$_2$O$_8$, 1.81(4) $\mu_B$, is considerably lower than that (∼ 2.2 $\mu_B$) in BaCo$_2$V$_2$O$_8$.  

FIG. 6. The horizontally summed intensity versus vertical pixel number for the (2, 3, δl) reflection as measured at 120 mK. (a) Rocking curves of the (2, 3, 0) reflection at 120 mK for $\mu_0 H \geq 12.0$ T.

FIG. 7. (a) Magnetic field dependence of the summed intensity of diffraction from the central analyzer blade on RITA-II (the area enclosed by the central white frame in Fig. 3a) measured at 120 mK. (b) Rocking curves of the (2, 3, 0) reflection at 120 mK for $\mu_0 H \geq 12.0$ T.
FIG. 8. (a) Magnetic field dependence of the FWHM of the (2, 3, 0) reflection at 120 mK measured in a \( h \)-scan on RITA-II. The green shaded area marks the LSDW region. (b) Temperature dependence of the (2, 0, 0) and (2, 3, 0) reflections at 7.5 T. A scale factor of 40 has been applied to the (2, 3, 0) reflection. The green arrows mark the transition temperatures described in the main text.

This could reflect the difference in the longitudinal dynamical structure factor \( S_{zz}(Q, \omega) \) in these two compounds. Empirically, \( S_{zz}(Q, \omega) \) depends on the magnetic anisotropy and interactions between the chains. The magnetic anisotropies in these two compounds are expected to be very similar, as supported by the similar values of the Landé \( g \)-tensor and anisotropic parameter \( \epsilon \) (Eq. 1). On the other hand, the interchain couplings are very complicated in relevant systems with the same crystallographic structure, which could afford exchange frustration. More importantly, it has been found that a mean-field theory is insufficient to properly reproduce the experimentally observed \( S_{zz}(Q, \omega) \) in SrCo\(_2\)V\(_2\)O\(_8\), necessitating the inclusion of all the interchain coupling terms for an accurate description.

One unique signature of the LSDW order in SrCo\(_2\)V\(_2\)O\(_8\) is the discrepancy between the theory and experiment in determining the value of the incommensurate modulation vector along the \( c \)-axis (\( \delta l \)) (Fig. 5a). In Section \( \gamma \) \( g_c \) and \( M_z \) measured between 1.3 and 1.9 K were used to estimate the theoretical values using Eq. 2. Interestingly, our measurements on the ferromagnetic reflection (2, 0, 0), which directly probes \( M_z \), clearly reveal a drop below \( \sim 0.3 \) K (Fig. 8b). This suppression is independent of the emergent AFM order, as the latter sets in at 0.6 K. Although we did not measure this reflection in the LSDW region due to the much weaker signal (0.1-0.2 \( \mu_B/Co \)), we believe that the suppressed \( M_z \) is responsible for the discrepancy shown in Fig. 5a.

In a TLL with Ising-like anisotropy, the critical magnetic field (\( H_c \)) at which the crossover between the longitudinal and transverse spin fluctuations occurs scales linearly with the intrachain exchange strength (\( J \) in Eq. 1). The LSDW order in an Ising-like quasi 1D quantum magnet results from the condensation of the longitudinal mode while the interactions between the chains become energetically relevant. An important conclusion revealed by the mean-field theory is that the collapse of the LSDW order does not coincide with \( H_c \), but shifts to a higher value. Therefore, the LSDW order is expected to be more robust in a system with a larger \( J \). Based on the high field magnetization and inelastic neutron scattering investigations, \( J \) is larger in SrCo\(_2\)V\(_2\)O\(_8\). However, the LSDW order in SrCo\(_2\)V\(_2\)O\(_8\) turns out to be more fragile (Fig. 7a). We note that the mean-field theory in Ref. 23 already fails to correctly reproduce the critical field for the LSDW to transverse AFM order transition in BaCo\(_2\)V\(_2\)O\(_8\). Our results further stress the inadequacy of the interchain mean-field theory in addressing the complex magnetism in these systems.

Our results suggest that the emergent AFM order sets in around 7.0 T, at which we performed two additional measurements with a much longer counting time on RITA-II. We could resolve a signal at 100 mK; this signal fades away at 750 mK. The reemergence of the scattering signal above 7.0 T (Fig. 7) is consistent with the behaviour of the transverse AFM order observed in BaCo\(_2\)V\(_2\)O\(_8\). However, the signal at 7.0 T is too weak...
to check the coexistence of the TAF and LSDW order reported in BaCo$_2$V$_2$O$_8$.$^{23}$

VIII. SUMMARY

In conclusion, we have employed single-crystal neutron diffraction to investigate the spin states in SrCo$_2$V$_2$O$_8$ up to 14.9T and down to 50mK (Fig.9). This system is composed of weakly coupled $S = 1/2$ XXZ spin chains with Ising-like anisotropy. Like its isostructural counterpart BaCo$_2$V$_2$O$_8$, this compound shows multiple magnetic-field-driven phase transitions that reflect the quantum critical nature of the spins in a TLL state. In addition to the similarity, we have identified several unique signatures for the spin states in SrCo$_2$V$_2$O$_8$, including the lower value of ordered magnetic moment, fragility of LSDW order, loss of three-dimensional long-range spin correlation in the LSDW region and suppression of uniform magnetization along the c-axis at low temperatures.

Our observations highlight the complex origin of the magnetism in these systems and evidence the inadequacy of the interchain mean-field theory. Further investigations, e.g. density functional theory calculations and inelastic neutron spectroscopy measurements, are in high demand to shed more lights on the interchain couplings in SrCo$_2$V$_2$O$_8$.

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