

## Kagome staircase compound $\text{Co}_3\text{V}_2\text{O}_8$ in an applied magnetic field: Single-crystal neutron diffraction study

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The magnetic properties of  $\text{Co}_3\text{V}_2\text{O}_8$  have been studied by single-crystal neutron diffraction. In zero magnetic field, the observed broadening of the magnetic Bragg peaks suggests the presence of disorder both in the low-temperature ferromagnetic and in the higher temperature antiferromagnetic state. The field dependence of the intensity and position of the magnetic reflections in  $\text{Co}_3\text{V}_2\text{O}_8$  reveals a complex sequence of phase transitions in this Kagome staircase compound. For  $H\parallel a$ , a commensurate-incommensurate-commensurate transition is found in a field of 0.072 T in the antiferromagnetic phase at 7.5 K. For  $H\parallel c$  at low temperature, an applied field induces an unusual transformation from a ferromagnetic to an antiferromagnetic state at about 1 T accompanied by a sharp increase in magnetization.

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### I. INTRODUCTION

The family of transition-metal vanadates  $M_3\text{V}_2\text{O}_8$  with  $M$  being Co, Ni, Cu, and Mn has been intensively studied in the context of frustrated magnetism<sup>1-5</sup> since the magnetically coupled ions in these compounds form a network reminiscent of the Kagome lattice. The chemical bonding between the magnetic  $M^{2+}$  ions (the  $\text{V}^{5+}$  ions remain nonmagnetic) forces the magnetic layers to buckle into a staircase formation, the so-called Kagome staircase lattice. Although the magnetic properties of different members of the family vary significantly, a unifying feature for all the members is an extreme richness of their highly anisotropic magnetic phase diagrams.<sup>5-7</sup> The abundance of magnetic phases is attributed to a close proximity in energy of several competing states with the selection of the ground state being readily influenced by a subtle balance among the nearest- and further-neighbor exchange interactions, magnetic anisotropy, and an applied magnetic field. An additional interest in  $\text{Ni}_3\text{V}_2\text{O}_8$  is associated with the discovery of magnetically driven ferroelectric order in this compound<sup>2</sup> while studies of the magnetic ordering in  $\beta\text{-Cu}_3\text{V}_2\text{O}_8$  with  $S=1/2$  are justified by the possible influence of quantum effects.<sup>3</sup>

In  $\text{Co}_3\text{V}_2\text{O}_8$  (CVO) a magnetic ordering at 11.3 K is seen as a relatively small peak in the heat capacity and a small kink in the magnetic susceptibility<sup>1</sup> while a further magnetic transition at 6 K is accompanied by a much more pronounced peak in the  $C(T)$  curve and a significant rise in  $\chi(T)$ .<sup>1</sup> By using neutron diffraction measurements, Chen *et al.*<sup>4</sup> have shown that the transition at 11.3 K is from a paramagnetic to an antiferromagnetic (AFM) incommensurate state and that the transition at 6 K is to a ferromagnetic (FM) state. A number of lock-in transitions associated with incommensurate-to-commensurate phases have also been observed at intermediate temperatures.<sup>4</sup> A peculiar feature of the FM state is that the size of the moment on one of the two  $\text{Co}^{2+}$  sites, the so-called cross-tie site, is considerably reduced compared to the fully polarized state.<sup>4,8</sup> The application of a magnetic field in this phase is found to enhance rapidly the cross-tie site magnetic moment, which reaches the expected value of  $3 \mu_B$  in higher fields.<sup>8</sup>

In an applied magnetic field, CVO demonstrates highly anisotropic behavior both in terms of the absolute values of susceptibility<sup>9</sup> and in terms of the sequence of the field-induced phase transitions.<sup>6,10</sup> Although the previous reports based on different experimental methods agree, in general, they disagree over the detailed description of the behavior of CVO in an applied magnetic field and, in particular, on the interpretation of the different magnetic phases.<sup>6,10-14</sup>

Despite the recent progress in probing the magnetic interactions by inelastic neutron scattering<sup>15</sup> a unified view of the strength and signs of all the relevant magnetic forces has yet to be reached, as none of the proposed models is capable of explaining the sequence of phase transitions observed in CVO. This absence of a generally accepted model is not particularly surprising, as a full description should incorporate 12  $\text{Co}^{2+}$  ions on two different sites in the unit cell (see Fig. 1) in the presence of significant orbital effects (the rather small variation of energy with  $q$  of the observed spin waves compared to the gap suggests that the strength of exchange interactions in CVO is comparable with the single-ion anisotropy<sup>16</sup>). The matter is complicated further by the possibility of nonzero field-induced moments on the vanadium and oxygen ions.<sup>17</sup>

We have performed single-crystal neutron diffraction measurements for two different directions of an applied magnetic field. For  $H\parallel a$ , an unusual transition is found at intermediate temperatures. For  $H\parallel c$ , the observation of a field-induced AFM state implies a significant influence of the inter-Kagome-plane interactions on the magnetic structure formation. The measurements have also shown that not all of the explored magnetic phases of CVO are long range in their nature and that a significant degree of magnetic disorder is often present.

### II. EXPERIMENTAL PROCEDURE

A CVO single-crystal sample of size  $4 \times 2 \times 2 \text{ mm}^3$  was prepared as described previously.<sup>9</sup> Single-crystal diffraction was performed on the D10 diffractometer at the Institut Laue-Langevin, Grenoble, France. An  $80 \times 80 \text{ mm}^2$  two-

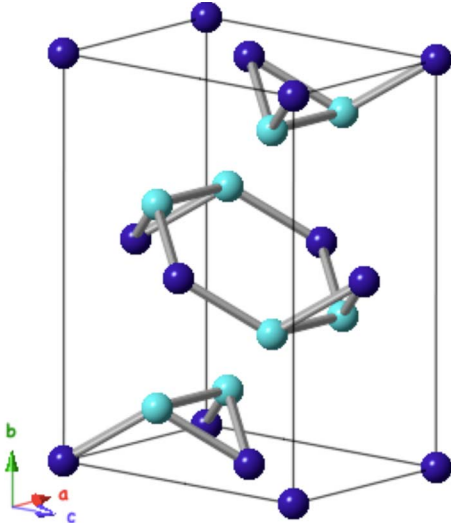


FIG. 1. (Color online) The positions of the magnetic  $\text{Co}^{2+}$  ions in the orthorhombic unit cell of  $\text{Co}_3\text{V}_2\text{O}_8$  (space group  $Cmca$ ). The cross-tie sites are shown in dark blue and the spine sites are in light blue.

dimensional microstrip detector was used in the diffraction configuration. Measurements were performed with  $\lambda = 1.53 \text{ \AA}$  and  $\lambda = 2.36 \text{ \AA}$ . A pyrolytic-graphite monochromator and filter were used for the longer wavelength while for the shorter wavelength a  $\text{Cu}(200)$  monochromator was used, still with the graphite filter to reduce the small half-wavelength contamination. A vertical magnetic field of up to 2.5 T supplied by a standard cryomagnet was applied either along the  $a$  axis or along the  $c$  axis limiting the observable scattering to the planes  $(0kl)$  and  $(hk0)$ , respectively. The measurements were performed by either ramping the magnetic field at constant temperatures and summing up the counts in a small area of the detector surrounding the reflection to obtain its peak intensity or by making  $\omega$  scans at various fixed values of field/temperature.

### III. EXPERIMENTAL RESULTS

#### A. Zero field

The temperature evolution of the magnetic ordering in CVO was initially studied by collecting the intensity of the  $(0k2)$  scans at different temperatures in zero field. In general agreement with previously reported single-crystal<sup>4,10</sup> and powder<sup>8</sup> neutron diffraction data, the sequence of the magnetic transitions on cooling in zero field is from a paramagnetic phase to a higher temperature AFM incommensurate phase (around 11.3 K), to a commensurate AFM  $k = \frac{1}{2}$  phase (just below 9 K), to a lower temperature AFM incommensurate phase (just above 7.5 K), to a commensurate AFM  $k = \frac{1}{3}$  phase (stable in a narrow region between 6.1 and 5.8 K) and, finally, to a FM phase, which is stable below 5.8 K.

More accurate measurements of the positions and shapes of the magnetic reflection performed at different temperatures with an analyzer in place of the position sensitive detector are shown in Fig. 2. In the paramagnetic region, at  $T$

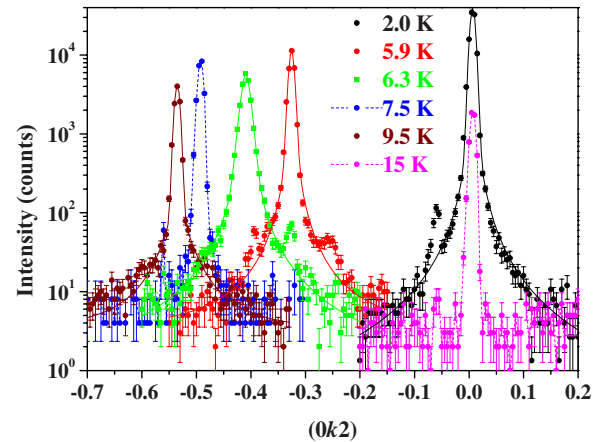


FIG. 2. (Color online) Temperature dependence of neutron diffraction profiles of scans along the  $(0k2)$  direction in zero field. The measurements were performed with the use of a vertically focusing pyrolytic-graphite analyzer. Solid lines are the fits consisting of two components, a main narrow Gaussian component and a less intense, broad Lorentzian component. Apart from the peak at  $T = 6.3 \text{ K}$  the Gaussian component is resolution limited. The data at 7.5 and 15 K are not fitted, as they have only the Gaussian resolution limited component.

$= 15 \text{ K}$ , the  $(002)$  peak has a nearly perfect Gaussian shape (apart from a small satellite, less than 0.3% of the intensity of the main peak), which is limited by the instrumental resolution. The same peak in the FM region has pronounced non-Gaussian tails, which amount to approximately 10% of the integrated intensity (see Fig. 2) and imply the presence of magnetic disorder throughout the sample. The incommensurate peak at 9.5 K and the  $k = \frac{1}{3}$  AFM peak at 5.9 K also exhibit a dominant resolution-limited component and a much broader component, which could be adequately approximated by a Lorentzian shape, although very small additional reflections on top of the Lorentzian cannot be completely ruled out. The main component of the incommensurate AFM peak at  $T = 6.3 \text{ K}$  is not resolution limited, its width is estimated as  $0.0151(5)$  r.l.u. along the  $(0k2)$  direction, which indicates that the magnetic correlation length along the  $b$  axis does not exceed  $220 \text{ \AA}$  at this temperature.

The presence of a significant short-range component in magnetic order (manifesting itself as broad peaks underneath the resolution-limited Bragg peaks) has been reported in other frustrated magnetic materials, ranging from an XY pyrochlore system  $\text{Er}_2\text{Ti}_2\text{O}_7$  (Ref. 18) to an Ising-type triangular antiferromagnet  $\text{Ca}_3\text{Co}_2\text{O}_6$ .<sup>19,20</sup> Although the mechanism responsible for the appearance of a short-range component is different in these compounds, the unifying motif is apparent: in the presence of frustration, the magnetic ground states corresponding to drastically different arrangements of magnetic moments can become energetically nearly equivalent. Therefore transitions between them often result in less than perfect order, especially at low temperatures.

#### B. Field parallel to $a$ axis

The measurements of the peak intensity as a function of magnetic field and the  $\omega$  scans both show that at the base

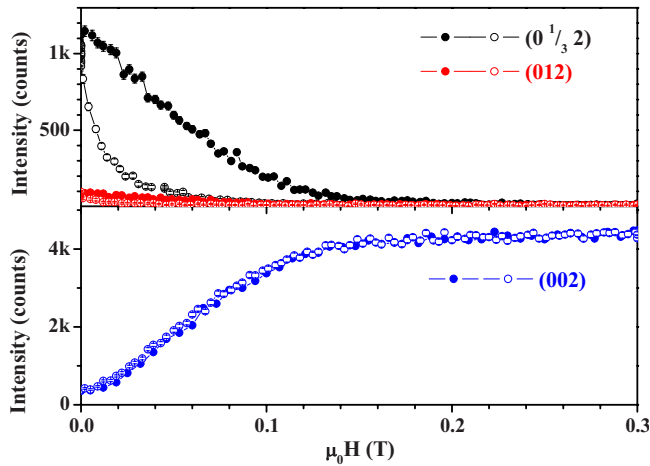


FIG. 3. (Color online) Magnetic field dependence of the integrated intensity of representative AFM (top panel) and FM (bottom panel) peaks in  $\text{Co}_3\text{V}_2\text{O}_8$  at  $T=6$  K for  $H\parallel a$ . Solid/open symbols denote increasing/decreasing magnetic field ramps.

temperature of 1.6 K, the application of a magnetic field of up to a maximum of 2.5 T along the  $a$  axis does not significantly change the intensity of the FM peaks. No new peaks have been found in applied magnetic fields along the  $(0k1)$  and  $(0k2)$  lines in reciprocal space. Neither the position, nor the shape, nor the intensity of the FM peaks such as, for example,  $(002)$  and  $(022)$  are altered by an applied magnetic field. This observation suggests that the rapid increase in magnetization seen at base temperature in small fields (up to 0.1 T) (Ref. 6) is caused by a rearrangement of magnetic domains. However, the measured value of the saturated magnetic moment, of about  $3.5 \mu_B$  per  $\text{Co}^{2+}$  ion, is significantly larger than the average of the zero-field-ordered magnetic moments on spine and cross-tie sites, which are only  $2.73 \mu_B$  and  $1.54 \mu_B$  according to powder neutron diffraction reports by Chen *et al.*<sup>4</sup> or  $3.04 \mu_B$  and  $1.81 \mu_B$  according to Wilson *et al.*<sup>8</sup> giving indirect support to the idea of nonzero moments on the vanadium and oxygen ions.<sup>17</sup> The earlier reported gain in the intensity of the FM peaks (and the associated values of the magnetic moment on the cross-tie site) in a powder neutron diffraction experiment<sup>8</sup> is much more gradual in nature, as saturation is reached at about 8 T.

At  $T=6$  K, which in zero field corresponds to the  $k=\frac{1}{3}$  AFM state, the field dependence of the magnetic peaks demonstrates a steady increase in intensity for the FM type and a gradual intensity decrease for the AFM type, as can be seen in Fig. 3. An obvious conclusion is that at this temperature, a relatively weak field, less than 0.2 T, destroys the AFM order and induces the FM state. Remarkably, a significant hysteresis is observed in the intensity of the AFM peaks (Fig. 3, top panel) while the intensity curves for the FM peaks for increasing/decreasing field are practically indistinguishable (Fig. 3, bottom panel).

On measuring the peaks at  $T=7.5$  K (see Fig. 4), two field-induced phase transitions have been found. The transition at 0.2 T is marked by the disappearance of the AFM peaks and also by a sharp increase in the intensity of the FM peaks, which then remain constant up to 2.5 T. This transition, accompanied by significant hysteresis, is from the AFM

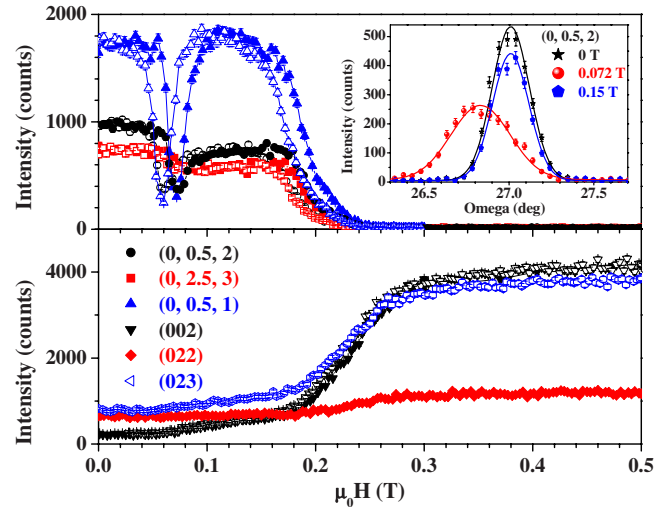


FIG. 4. (Color online) Magnetic field dependence of the integrated intensity of several AFM (top panel) and FM (bottom panel) peaks in  $\text{Co}_3\text{V}_2\text{O}_8$  at  $T=7.5$  K for  $H\parallel a$ . Solid/open symbols denote increasing/decreasing magnetic field ramps. The inset shows  $\omega$  scans for the  $(0\frac{1}{2}2)$  reflection at 7.5 K in zero field, 0.072 and 0.15 T. Solid lines are Gaussian fits. The full width at half maximums of the peaks are  $0.26^\circ$ ,  $0.41^\circ$ , and  $0.25^\circ$ , respectively.

to the fully polarized FM state, where a nearly full magnetic moment on both  $\text{Co}^{2+}$  sites is recovered according to the magnetization measurements.<sup>6</sup>

Another transition in a lower field ( $\approx 0.072$  T with a significant hysteresis) is clearly marked by a sharp minimum in the intensity of the AFM peaks, as shown in Fig. 4, top panel. The  $\omega$  scans performed on the AFM reflection  $(0\frac{1}{2}2)$  at 7.5 K in zero field, 0.072 and 0.15 T have revealed, however, that the observed intensity decrease is mostly due to the movement of the peak to a slightly different position (see inset in Fig. 4). A shift of approximately  $0.18^\circ$  away from the  $(0\frac{1}{2}2)$  position suggests that in a field of 0.072 T the magnetic structure is incommensurate with  $k \approx 0.475$  instead of the commensurate AFM position with  $k = \frac{1}{2}$ , although a considerable increase in the peak width implies that there is a large spread in the value of  $k$  at the transition point. The transition at 0.072 T is also marked by a small change in the slope of the intensity of the FM peaks (see Fig. 4) and by a change in slope in the magnetization versus magnetic field.<sup>6</sup>

### C. Field parallel to $c$ axis

In sharp contrast to the results described above for  $H\parallel a$ , the application of a magnetic field along the  $c$  axis is accompanied by a pronounced change in intensity of the magnetic peaks at all temperatures, including at the base temperature of 1.6 K. Figures 5 and 6 show the field dependence of the intensity of magnetic peaks at  $T=1.6$  K and  $T=7.5$  K, respectively.

At the lower temperature, a pronounced phase transition is seen just below 1 T. The transition is characterized by an abrupt gain in the intensity of such peaks as  $(020)$ ,  $(030)$ ,  $(060)$ ,  $(220)$ , and  $(420)$  at the expense of such peaks as  $(040)$  and  $(440)$ . The intensities of the  $(020)$  and  $(220)$  peaks have



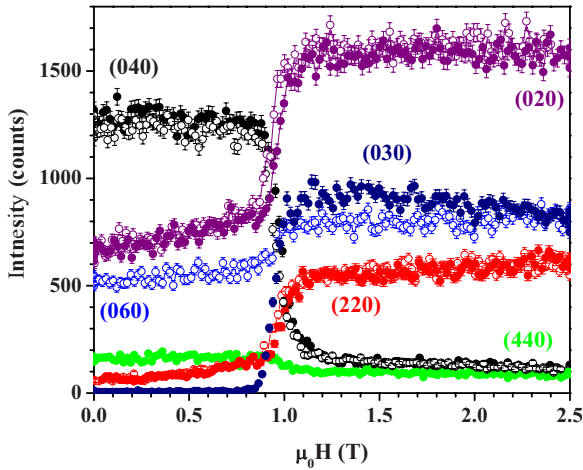


FIG. 5. (Color online) Magnetic field dependence of the integrated intensity of several peaks in a single crystal of  $\text{Co}_3\text{V}_2\text{O}_8$  at  $T=1.6$  K for  $H\parallel c$ . Solid/open symbols mark increasing/decreasing magnetic field ramps.

a noticeable positive slope in lower fields, which is compatible with the steady rise of the magnetization.<sup>6</sup> This gradual increase in intensity corresponds to a deviation of the magnetic moments away from their initial orientation parallel to the  $a$  axis toward the magnetic field direction along the  $c$  axis. The intensity of the (030) peak is zero in lower fields followed by a sharp increase just below 1 T (see Fig. 5). The field variation of the intensity of this peak has a small but distinctly negative slope above 1 T, which indicates that the magnetic structure in this field range has a sizeable antiferromagnetic component and that this component is decreasing steadily in an increasing field, as the magnetic moments become more and more polarized along the field.

The transition into an AFM phase from the initial FM ground state is found to be temperature sensitive, as it shifts

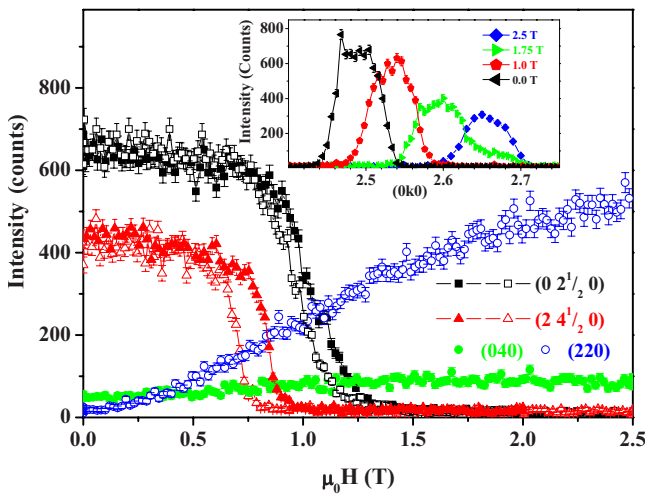


FIG. 6. (Color online) Magnetic field dependence of the integrated intensity of several AFM and FM reflections in a single crystal of  $\text{Co}_3\text{V}_2\text{O}_8$  at  $T=7.5$  K for  $H\parallel c$ . Solid/open symbols mark increasing/decreasing magnetic field ramps. The inset shows the magnetic field dependence of the neutron diffraction profiles taken along the  $(0k0)$  direction at  $T=7.5$  K.

from just below 1 T at 1.6 K to nearly 2 T at  $T=5.8$  K. The maximum experimentally available field of 2.5 T has not permitted the observation of a further transition to a fully polarized state, which takes place at 5 T for  $T=2$  K,  $H\parallel c$ .<sup>6</sup>

Unlike the intermediate-temperature zero-field AFM structure characterized by the propagation vector  $k=(0\frac{1}{2}0)$ , the low-temperature field-induced AFM structure does not involve a doubling of the unit cell. The observed (030) AFM reflection corresponds to the propagation vector  $k=(010)$  [or  $(\frac{1}{2}\frac{1}{2}0)$  in a primitive cell] implying that the  $\text{Co}^{2+}$  atoms located in the adjacent Kagome planes carry magnetic moments with significant antiparallel components. This in turn indicates that the exchange interaction between the planes is strong and antiferromagnetic in nature. This interaction has so far been neglected in the models proposed to describe the inelastic neutron scattering results.<sup>15</sup> Although the observed field-induced transition is certainly of the FM to AFM type, a full refinement of the AFM structure above this transition was not possible due to a very limited number of magnetic reflections collected.

At  $T=7.5$  K the intensity of the integer FM peaks (see Fig. 6) is a smooth function of the applied field in the same manner as the overall magnetization of the sample.<sup>6</sup> The field dependence of the intensity of the AFM half-integer peaks such as  $(0\frac{5}{2}0)$  and  $(2\frac{9}{2}0)$ , however, still demonstrates a pronounced change around 1 T. This abrupt change in intensity (which is accompanied by a significant hysteresis) corresponds to the change in position of the AFM reflections in reciprocal space rather than to their complete disappearance. The scans taken along the  $(0k0)$  direction in different fields (see inset to Fig. 6) show that in higher fields the magnetic structure is incommensurate and the peaks are seen at general positions  $(h, \frac{1}{2} + \delta, l)$ , where  $0 < \delta < 0.15$ , rather than at half integer AFM positions  $(h\frac{1}{2}l)$ .

These observations encourage an analogy between the effects caused by an applied magnetic field and increasing temperature in  $\text{Co}_3\text{V}_2\text{O}_8$ . In zero field, a temperature increase initially induces a transition from the FM ground state to an AFM phase and then to an incommensurate phase. The effects of applying a magnetic field along the  $c$  axis are not dissimilar, as it causes the transition from the FM phase to the AFM phase at lower temperatures and a transition from an AFM state to an incommensurate state at higher temperatures.

There is an interesting possibility to consider here. As much as the complex competing interactions are likely to be responsible for the appearance of a large number of magnetic phases in  $\text{Co}_3\text{V}_2\text{O}_8$  in an applied field, the interactions themselves could be modified significantly by the transitions through the local lattice distortions involving Co center displacements.<sup>21</sup>

IV. SUMMARY

Single-crystal neutron diffraction studies of the magnetic properties of the Kagome staircase compound  $\text{Co}_3\text{V}_2\text{O}_8$  have been performed in zero field and also for two different directions of an applied field,  $H\parallel a$  and  $H\parallel c$ . Zero-field data have revealed the presence of magnetic disorder at different tem-

peratures, particularly identifiable in the antiferromagnetic phase around  $T=6.3$  K, for which the magnetic correlation length is limited to  $220 \text{ \AA}$ . For  $H\parallel a$ , an unusual transition involving a shift of the magnetic Bragg peaks from an antiferromagnetic to an incommensurate and back to an antiferromagnetic position has been observed. For  $H\parallel c$ , the measurements have allowed for identification of the observed magnetic field-induced phase as antiferromagnetic, despite the sharp increase in the magnetization at a transition point. Our results reveal a need for the development of a theoretical

model which will include the interplanar exchange interactions and call for further inelastic neutron-scattering experiments, which can probe these interactions directly.

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