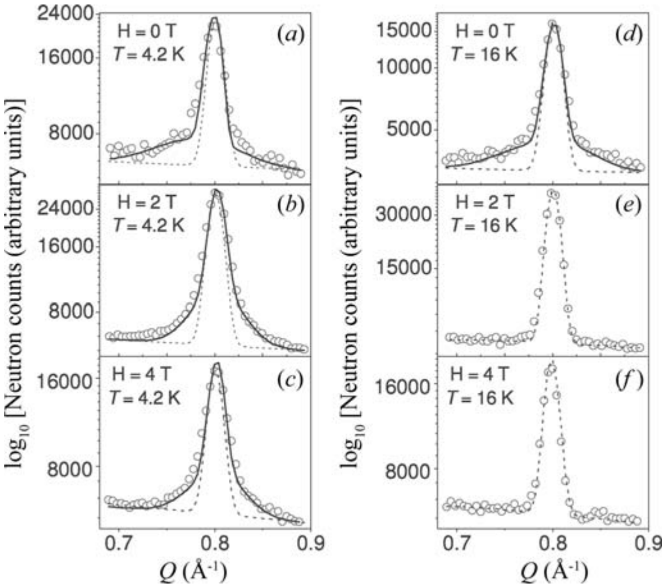

Figure 2.8.15
The observed Bragg reflection 100 (open circles) under an applied field of (*a*) 0 T, (*b*) 2 T and (*c*) 4 T at 4.2 K and (*d*) 0 T, (*e*) 2 T and (*f*) 4 T at 16 K (taken from Yusuf *et al.*, 2013). Copyright IOP Publishing. Reproduced with permission. All rights reserved.

narrow temperature range a macroscopic polar vector leads to a multiferroic behaviour. As this study was based on single-crystal neutron measurements, no further details are given here. Frustrated triangular-lattice Ising antiferromagnets have degenerate magnetic ground states, which give rise to very complex magnetic structures. As there are only small differences in the competing exchange interaction in such frustrated triangular-lattice compounds, a sequence of phase transitions is introduced by changes in temperature or magnetic field. The compound $Ca_3Co_2O_6$ is another example of a frustrated system. Field-dependent powder diffraction patterns were reported for the doped system $Ca_3Co_{1.8}Fe_{0.2}O_6$ by Yusuf *et al.* (2013). They distinguished the short-range magnetic order (SRO), reflected in the half-width of the Bragg reflections (Fig. 2.8.15), from the long-range order as given by the Bragg positions. They stated that even under magnetic fields up to 4 T the broadening of Bragg reflections indicates the persistence of SRO. In a field of 2 T, the observed change in the structure from incommensurate to commensurate indicates a reduction of spin frustration. In fields of 4 T, a ferrimagnetic system is introduced, followed by a ferromagnetic one above 5 T.

2.8.3.3.2.2. *Manganite systems*

Like the vanadates, in the class of rare-earth manganites of the type RMn_2O_5 successive magnetic phase transitions between commensurate (CO) and incommensurate phases (ICP) can occur. Intensive investigations have been undertaken to understand the relationship between their magnetic and dielectric properties. The spontaneous electric polarization is induced by a magnetic transition. Thus the primary order parameter is magnetic rather than structural. Among the rare-earth compounds, those containing Nd or an element lighter than Nd do not exhibit ferroelectricity. In all these materials a broken magnetic symmetry at lower temperatures leads to a polar symmetry group. In addition, a cycloidal component indicates a common underlying mechanism. The Mn^{3+} and Mn^{4+} ions are fully charge-ordered. Neutron diffraction studies of these phases have been performed by Radaelli & Chapon (2008), who also analysed the possible exchange pathways. In $TbMn_2O_5$ the *H*–*T* phase diagram of the commensurate–low-temperature–incommensurate (CO–LT–ICP) magnetic transitions shows an upward jump in the transition temperature from ~25 K at zero field to 27 K at 9 T. The low-temperature ICP phase is stabilized under an external field for $TbMn_2O_5$ and the dielectric constant is enhanced. It was concluded that Tb and Mn order independently, implying the absence of coupling terms between them. Strong support for this suggestion was provided by an in-field neutron study on the analogue YMn_2O_5. Neither the positions nor the intensities of the magnetic Bragg reflections were affected by the magnetic field (Fig. 2.8.16). The magnetic low-temperature ICP phase in the Tb compound was stabilized under a magnetic field. This is in contrast to observations on $HoMn_2O_5$ by Kimura *et al.* (2007), using single crystals. In both cases, however, the neutron data correlate directly with the results obtained by dielectric measurements under a magnetic field. The difference in the behaviours is thus confirmed. The two studies also reveal different magnetic order at low temperatures. The same magnetic sequence at low temperatures as for Tb was observed in YMn_2O_5, which does not contain a magnetic rare-earth element. Under fields up to 8 T the positions and the intensities of the magnetic Bragg reflections remained unchanged, showing that the antiferromagnetic structure of the manganese sublattice is extremely stable. As in the vanadates, the main reason for the sequence of magnetic structures is frustration of the manganese spins. Without going too deeply into the details of the different exchange pathways and orbital occupancies, one factor behind this behaviour is the Jahn–Teller effect of the Mn^{3+} ion, which is also relevant in the multiferroic $TbMnO_3$ as part of the $RMnO_3$ family (Kimura *et al.*, 2003). Another feature often found in multiferroic systems is the small ferromagnetic component caused by small spin canting due to Dzyaloshinskii–Moriya interactions. This property strongly influences the low-temperature magnetism in RMn_2O_5 (Kimura *et al.*, 2009).

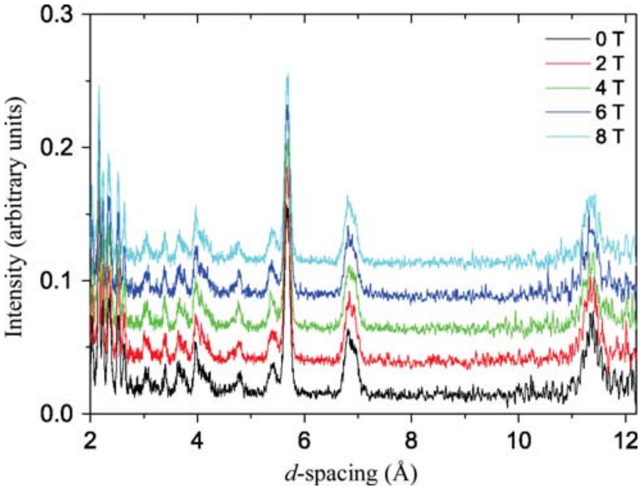

Figure 2.8.16
Time-of-flight diffraction patterns of YMn_2O_5 at 1.6 K under magnetic fields between 0 and 8 T (taken from Radaelli & Chapon, 2008). Copyright IOP Publishing. Reproduced with permission. All rights reserved.

2.8.3.3.3. *Additional systems and scattering techniques*

Information about the anisotropy of the local magnetic susceptibility at different magnetic sites has been extracted from diffraction patterns for a $Tb_2Sn_2O_7$ powder measured using polarized neutrons under magnetic fields of 1 and 5 T (Gukasov

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