Spin-reorientation in GdGa

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The orthorhombic RGa (R = rare earth) intermetallic compounds crystallize in the CrB-type *Cmcm* (#63) structure with one R site and one Ga site, both 4*c*. The RGa compounds were first studied in the early 1960s[1–4] and were found to order ferromagnetically with a Curie temperature ranging from a high of ∼ 187 K in GdGa to a low of 15 K for TmGa[5–11]. The magnetic structures of TbGa[12], ErGa[13], and HoGa[14] have been previously determined by neutron diffraction. However, as is usually the case for Gd-containing compounds, there had previously been no neutron diffraction measurements of GdGa due to the extremely large neutron absorption cross-section of Gd.

Within the last few years, the RGa compounds have attracted some interest due to their potential for use as magnetocaloric-effect-based low-temperature refrigeration materials[10,15–17]. Our interest in GdGa stems not only from the possible technological applications, but also from the ^{119}Sn Mössbauer work of Delyagin *et al.*[9]. They found that GdGa undergoes a spin-reorientation transition at T_{SR} = 85 K. In addition, while a single sextet was sufficient to fit the ^{119}Sn spectra of Sn-doped GdGa between T_c and T_{SR}, below T_{SR} two equal-area sextets were required to fit the ^{119}Sn spectra. The two sextets were interpreted as indicating that the Gd 4*c* site splits into two magnetically-inequivalent sites with magnetic moments canted by 30(5)° and 64(5)° away from the crystallographic *a*-axis.

Our aim was to make a direct measurement of the ordering direction of the Gd moments above and below T_{SR} using neutron diffraction and ^{155}Gd Mössbauer spectroscopy. As natural gadolinium is the most powerful neutron absorbing element we used a large-area flat-plate sample holder to perform the neutron diffraction measurements. We have successfully used this technique several times to obtain good quality thermal neutron diffraction patterns of Gd-containing compounds. A full description of the flat-plate sample holder has been published in the Journal of Applied Crystallography[18].

In Figure 1 we show the neutron powder diffraction pattern of GdGa at 220 K, 110 K and 3.6 K. For the 110 K pattern, we found that the Gd magnetic moments order ferromagnetically along the *b*-axis with μ_{Gd} = 4.6(1) μ_B. Analysis of the 3.6 K pattern revealed that the four Gd magnetic moments in the *Cmcm* cell split into two (2:2) magnetically inequivalent groups, differing only in their orientations. The Gd moments were found to lie in the *bc*-plane, canted by 84(3)° and 46(4)° from the *b*-axis, respectively. The refined Gd moment is 6.7(4) μ_B, in good agreement with the expected value of 7 μ_B.

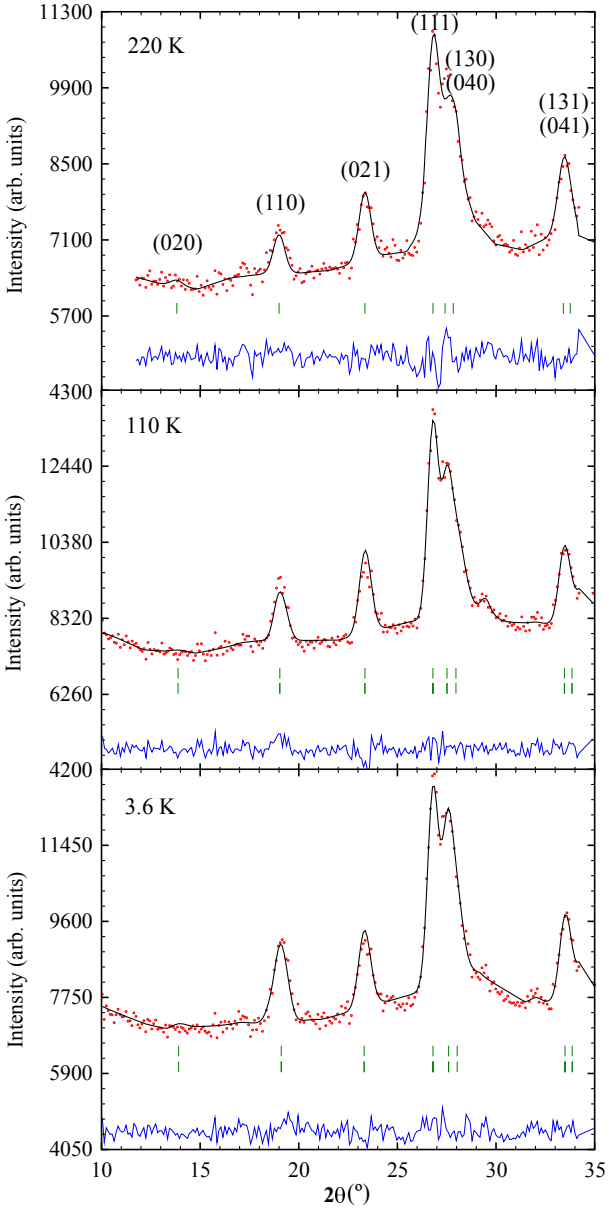

FIG. 1. Neutron powder diffraction patterns of GdGa obtained at 220 K, 110 K and 3.6 K, with a neutron wavelength of 1.3286(1) Å. The two sets of Bragg markers represent the nuclear and magnetic contributions of GdGa to the patterns. The residuals are shown in blue.

We also performed ^{155}Gd Mössbauer spectroscopy measurements to confirm the presence of two sites and their canting angles. Refinements of the Mössbauer spectra do indeed give two sites, with canting angles of 0(2)° and 49(2)° with respect to the principal axis of the elec-

tric field gradeient tensor (V_{zz}) at each site. If we select the correct electric field gradient axis → crystallographic axis combination then the results of the ^{155}Gd Mössbauer spectroscopy are in excellent agreement with the values obtained by neutron powder diffraction.

This study has revealed that GdGa does indeed order with two separate magnetic sites below T_{SR}. The magnetic moments are canted by 84(3)° and 46(4)° from the *b*-axis in the *bc*-plane. These results initially appeard to contradict those of Delyagin *et al.*, however we re-evaluated their ^{119}Sn Mössbauer spectra in light of our results and found that, with the proper EFG axis → crystallographic axis assignment, the 5 K pattern is consistent with the results of both neutron diffraction and ^{155}Gd Mössbauer spectroscopy. A complete description of this work has been published in the journal Hyperfine Interactions[19].

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