Magnetic Structure of Gd-Y Single-Crystal Alloys from Neutron Diffraction and Magnetization Measurements

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The first neutron-diffraction study of the helical phase of Gd-Y alloys in the vicinity of 30 at.% Y is presented. For each alloy composition the turn angle of the helix is seen to go smoothly to zero at the phase boundary with the low-temperature canted-ferromagnetic phase. The initial (onset) turn angle decreases rapidly with increasing Gd concentration, tending to zero at ~29.7 at.% Y. Combining the neutron data with magnetization, resistivity, and ultrasonic data allows us to construct a definitive magnetic phase diagram for the Gd-Y system. Two multicritical points are observed but neither complies fully with all the conditions necessary for a Lifshitz point.

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The Gd-Y alloy series has been the subject of considerable interest recently[1] in view of the competition between the ferromagnetic order favored by pure Gd and the helical magnetism found in the Y-rich alloys. Two multicritical points have been reported[2] in the region of 30 at.% Y where these two phases meet. The possible existence of a Lifshitz point near that composition has also been suggested.[3] A Lifshitz point, previously reported only for MnP,[4] occurs where three phases (paramagnetic, ferromagnetic, and a modulated structure with modulation wave vector $q \rightarrow 0$) meet at the triple point.

However, a complete understanding of this region of the magnetic phase diagram has been hampered by the lack of neutron scattering data due to the high capture cross section of gadolinium. The availability of high-intensity, low-wavelength neutrons has allowed the first comprehensive study of the magnetic structure of the Gd-Y alloys.

Here we present the results of the neutron study of the helical antiferromagnetic region close to 30 at.% Y, together with magnetization studies of samples cut from the same parent single crystals. These results, when supplemented with information from transport[2] and ultrasonic studies, allow us to present a definitive phase diagram. The existence of two multicritical points is confirmed, although neither fulfills the standard Lifshitz-point conditions.[3,5] In particular, the one observed at high gadolinium concentration is of a totally novel character.

The single crystals were grown in three separate centers; the Ames Laboratory, Iowa, The Clarendon Laboratory, Oxford, and the Centre for Materials Science, Birmingham. Both strain-anneal and float-zoning methods were used.

The neutron diffraction measurements were performed on the four-circle diffractometer D9 at the Institut Laue-Langevin, Grenoble. A wavelength of 0.52 Å was chosen to compromise between the increasing resolution and the rapidly increasing absorption cross section of gadolinium as the neutron wavelength is increased. The disk-shaped 31.1- and 30.4-at.%-Y samples were 6.3 and 5.2 mm in diameter and 2.4 and 3.9 mm thick, respectively, with the hexagonal *c* axis normal to the disk faces; the 32.0- and 30.0-at.%-Y samples were rectangular-box shaped with dimensions $4.2\times2.8\times2.8$ and $5.2\times4.2\times2.9$ mm^3, respectively. Scans along selected directions in reciprocal space were made over a temperature range of 170 to 230 K to detect possible antiferromagnetic satellite reflections near nuclear Bragg peaks. The samples were oriented with *b* ($\bar{1}20$ reciprocal-lattice direction) close to vertical. The scans were then done in the plane defined by the (100) and (002) reciprocal-lattice vectors primarily as scans from $(h,0,l-\epsilon)$ to $(h,0,l+\epsilon)$. The only satellites detected were of the type $h\ k\ l\pm\epsilon$, characteristic of basal-plane spirals.

The magnetic measurements were made with a vibrating-sample magnetometer described previously,[6] with samples cut with dimensions $3\times3\times5$ mm^3 with the *a*, *b*, or *c* axis along the long dimension, except for the *a*-axis crystal of 32 at.% Y which has dimensions of $1\times2.5\times4$ mm^3. The first magnetization curves were obtained in the residual field of the electromagnet (~38 Oe), and were inconsistent with measurements performed in the Earth's field alone. Hence, a reverse current was used in the electromagnet to give fields of about 8 Oe which proved to be very satisfactory except for occasional signal-to-noise problems.

Figure 1 shows the temperature dependence of the helical turn angle, θ, for the samples of 32, 31.1, 30.4, and 30 at.% Y calculated from the position of the satellites. No evidence of a helical regime was found for 29.7 at.% Y. The curves in Fig. 1 show a remarkable decrease of θ with decreasing temperature, leading virtually to a zero turn angle at the helical-ferromagnetic transition point (T_F). This suggests a second-order phase transition at T_F in contrast to the case of helimagnetism in Dy and Tb, for which pronounced discontinuities in θ are observed at the first-order transitions at T_F. Although the finite resolution does not allow us to probe all the way to $\theta = 0$, we have obtained a value of θ of 1.4° for 32 at.% Y. This is by far the smallest turn angle observed for a rare-earth material.

In addition, the onset turn angles are remarkably low in this composition range. The inset to Fig. 1 shows the initial turn angle (θ_i) of the various samples plotted against the de Gennes factor, where it is evident that a sample with just less than ~ 30 at.% Y would have a zero onset turn angle (i.e., no helical phase) as has been experimentally observed for 29.7 at.% Y.

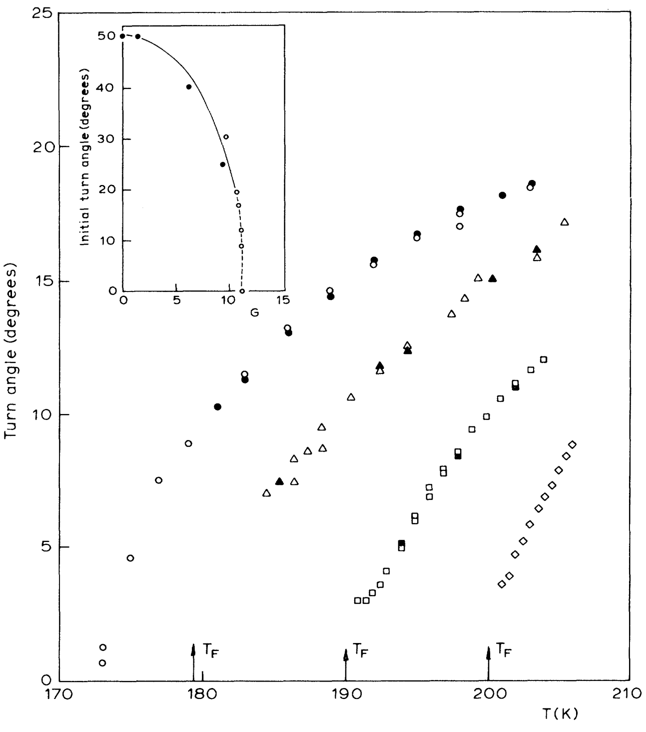

FIG. 1. Temperature dependence of the helical turn angle θ for 32.0, 31.1, 30.4, and 30.0 at.% Y in Gd single-crystal samples. Open and solid circles, $Gd_{68.0}Y_{32.0}$ cooling and warming, respectively; open and solid triangles, $Gd_{68.9}Y_{31.1}$ cooling and warming, respectively; open and solid squares, $Gd_{69.6}Y_{30.4}$ cooling and warming, respectively; lozenges, $Gd_{70.0}Y_{30.0}$ cooling for three different reflections (101, 102, 100). The inset shows the initial turn angle (turn angle on the T_N boundary of Fig. 3) plotted against the de Gennes factor (G). The solid line represents the results for other rare-earth metals and alloys; solid circles, Gd-Y data of Child and Cable (Ref. 7); open circles, present work.

It now appears that the magnetic measurements reported previously by Ito and co-workers[1] for the 30-at.%-Y alloy were obtained in a field (39 Oe) that suppressed the helical structure. The existence of such a phase in zero field is confirmed by the neutron observation of the helix satellites, by ultrasonic measurements of the elastic constant $C_{33}(T)$ and its associated ultrasonic attenuation $\alpha_{33}(T)$, and by the temperature derivative of the electronical resistivity $(d\rho/dT)$.[8]

The magnetization (σ) of c- and a-axis crystals of 30.5 at.% Y in applied fields of about 17.5 and 8 Oe is shown in Fig. 2 (upper curves). At high temperatures all curves show the same type of behavior, a sharp increase of σ as T is reduced towards T_C. *With a c-axis field,* the magnetization becomes constant below the Curie point $T_C = 214.8$ K, in a manner characteristic of a demagnetization-limited result. This is a clear confirmation of the initial ordering of a c-axis moment for this sample. At lower temperatures σ shows a rapid

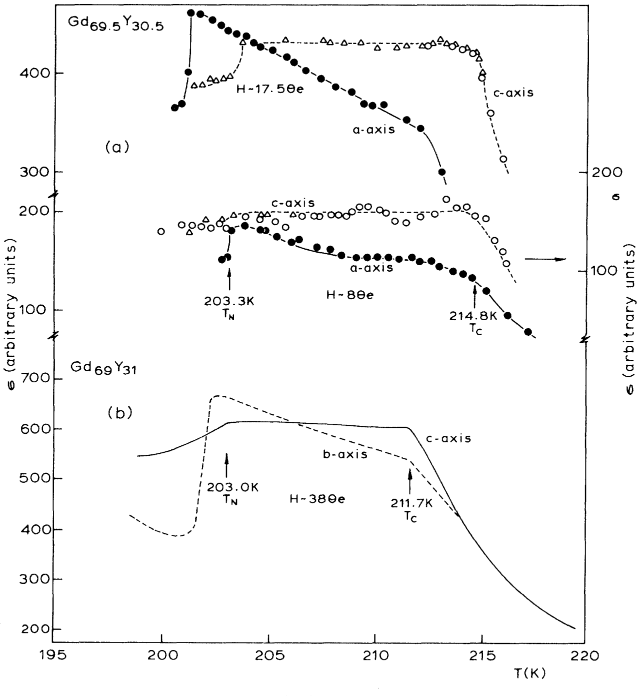

FIG. 2. Magnetization (σ) in arbitrary units vs temperature for (a) 30.5 at.% Y in Gd single-crystal samples in 8- and 17.5-Oe applied fields and for (b) 31 at.% Y in Gd single-crystal samples in a 38-Oe applied field.

drop, due to the sudden decrease in the *c*-axis moment as the sample orders in a basal-plane helical phase at $T_N = 203.3$ K. *With the field along the a axis,* anomalies mark both the onset and the extinction of the ferromagnetic phase, ferro I. However, as T decreases in this phase, no flattening off at a constant value is observed. Instead σ rises steadily with an upward curvature, indicating a continuing disorder in the basal plane. Since we have an ordered *c*-axis moment, the basal-plane results disprove the possibility of a canted ferromagnet, and support the model of a *c*-axis ordered moment with random-moment components in the basal plane.

We should point out that both T_C and T_N are hardly affected by a *c*-axis field, whereas a large effect is evident for a field applied parallel to the *a* axis. Increase of the basal-plane field seems to delay the onset of the *c*-axis ferromagnetic phase in this sample.

The magnetization curves for the 31-at.%-Y sample are shown in the lower part of Fig. 2, and they exhibit a behavior similar to that observed in $Gd_{69.5}Y_{30.5}$. T_C and T_N are 211.7 and 203.0 K, respectively, the extent of the ferro-I phase being reduced to 8.4 K with respect to the more Gd-rich alloy (11.5 K). The direction of the applied field does not appear to have a significant effect on T_C and T_N.

The magnetization results on 32- and 32.3-at.% single crystals yield $T_C = 210.3$, $T_N = 208.4$ K and $T_C = 208.4$, $T_N = 207$ K, respectively. Qualitatively they are very similar to the results on the samples of 30.5 and 31.0 at.% Y, except for the drastic reduction in the width of the ferro-I phase, $\Delta T = 1.9$ and 1.4 K for 32 and 32.3 at.% Y, respectively. It should be noted that there was little thermal hysteresis. Measurements in an 8-Oe applied field on 32.7 at.% Y revealed that this alloy orders directly in a helical structure at $T_N = 206.3$ K with no intermediate ferro-I phase. These data are not shown.

A phase diagram has been constructed for the Gd-Y system in the vicinity of 30 at.% Y and is shown in Fig. 3. The individual phase transitions were found by averaging results from neutron diffraction, ultrasonic, and resistivity experiments. The magnetization measurements described above have not been used since they were not taken in zero field; however, they are in close accord with Fig. 3. The phase diagram reveals some unusual features although overall it shows consistent results between the various experimental techniques. In the upper ferromagnetic region the *c* axis is the easy direction, while the lower ferromagnetic region has a canted ordered moment with the cant angle decreasing with yttrium concentration.[9] The phase boundaries T_N and T_F both show features characteristic of second-order changes. The two compositions marked A and B each exhibit a multicritical point. For the composition B the multicritical point occurs at a temperature of ~ 208 K and separates the paramagnetic, *c*-axis ferromagnet, and helical phases. However, this cannot be a Lifshitz point since the turn angle, θ, of the spiral is far from zero whereas at the Lifshitz point θ should tend to zero. In fact, the phase boundary labeled T_F is probably a line of $\theta \rightarrow 0$. The results shown in Fig. 1 indicate that 32-, 31.1-, 30.4-, and 30-at.%-Y samples all have turn angles that decrease smoothly to zero with decreasing temperature. In addition, very similar behavior has been observed for a 38-at.%-Y sample where the minimum measured value of θ was $\sim 1°$ and was tending to zero at T_F.[9,10]

We therefore have the intriguing situation that point A in the phase diagram separates three phases, viz., helical, canted ferromagnet, and *c*-axis ferromagnet with basal-plane disorder. The critical exponent of the initial turn angle along the line T_N has a value of 0.38, much lower than one would expect for a Lifshitz point.[3,5]

In summary, we now have a definitive phase diagram for the Gd-Y system in the vicinity of 30 at.% Y. It seems unlikely that either of the multicritical points observed are three-dimensional Lifshitz points although point A may well be a two-dimensional Lifshitz point. To investigate point A further we have prepared a 29.7-at.% sample and results will be presented shortly. Most importantly we have observed that, within the limits of resolution of our experiments, the helical–ferro-II phase boundary in Gd-Y marks a boundary where the propagation vector q (and therefore the turn angle of the spiral) of the modulated structure goes smoothly to zero.

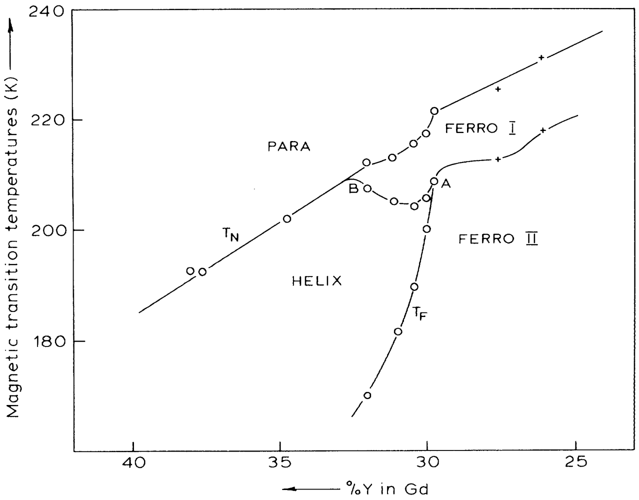

FIG. 3. Partial magnetic phase diagram for Gd-Y alloys showing magnetic ordering temperatures vs at.% Y in Gd in the range of 40 to 25 at.% Y.

(a)Deceased.

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