A CANTED MAGNETIC STRUCTURE IN AN S-ION COMPOUND: GdMg

P. MORIN, J. PIERRE and D. SCHMITT
Laboratoire Louis Néel, C.N.R.S., 166X, 38042 Grenoble-Cedex, France

and

D. GIVORD [1]
Institut Laue–Langevin, 156X, 38042 Grenoble-Cedex, France

Received 6 December 1977

A canted structure has been observed in GdMg by neutron diffraction at 0.5 Å. That suggests for a cubic S-ion compound the existence of additional exchange interactions besides the Heisenberg coupling.

Rare-earth metals form equiatomic compounds having the cubic CsCl-type structure with metals such as Rh, Cu, Ag, Zn or Mg [1]. They constitute a very simple system for the systematic study of the 4f magnetic interactions.

GdMg [2,3] orders at 110 K and bulk magnetic measurements indicate a spontaneous magnetization of 4.54 μ_B at 4.2 K, much lower than the 7 μ_B of the Gd^{3+} ion. Furthermore a large susceptibility is observed in magnetic fields up to 150 kOe, where the magnetic moment reaches only 6.22 μ_B. These results suggest that GdMg is not a simple ferromagnet.

The large absorption of natural gadolinium for thermal neutrons has previously prevented detailed neutron studies, except with non absorbing isotope [4]. However, the reduction in the absorption with smaller wavelength has allowed a partial single crystal investigation of Gd [5] and $Gd_{0.9}Y_{0.1}$ [6]. The cross section of natural gadolinium is only 250 barn at $\lambda = 0.5$ Å, a wavelength close to the peak neutron flux at the hot source of the Institut Laue–Langevin reactor in Grenoble (diffractometer D5). The flux at the specimen was about 2×10^6 neutrons cm^{-2} s^{-1}. In order to minimize the absorption further, the polycrystalline sample was a flat plate 1 mm thick and 25 cm^2 in surface. The experiments were performed at different temperatures from 12 K to 130 K. At the lowest temperature a magnetic field of up to 2 kOe was applied along the scattering vector $\boldsymbol{k}$, in the plane of the plate. The demagnetizing field measured by magnetization experiments was 400 Oe.

Fig. 1 shows the neutron diffraction patterns obtained at 130 K and 12 K. The 130 K spectrum is consistent with the coherent nuclear scattering expected from a pure cubic CsCl structure; in addition lines from the aluminium tails of the helium refrigerator are observed. Using the value of the nuclear scattering amplitude $b_{Mg} = 0.52 \times 10^{-2}$ cm, the best agreement with the experimental nuclear intensities was obtained for the value $b_{Gd} = (1.1 \pm 0.1) \times 10^{-12}$ cm. This value is in agreement with that of $b_{Gd} = 0.95 \times 10^{-12}$ cm obtained for $\lambda = 0.35$ Å [5].

At 12 K we observe simultaneously the growth of nuclear lines and the appearance of new reflections which are indexed as $(h, k, \frac{1}{2})$ in the cubic cell. This is consistent with the presence of both ferromagnetic and antiferromagnetic contributions. Evaluating separately each component and taking into account the magnetic form factor of Gd^{3+} [7] we obtain $\mu_F = 4.9 \pm 0.3$ μ_B and $\mu_{AF} = 5.1 \pm 0.3$ μ_B for the ferromagnetic and antiferromagnetic components respectively. Within the experimental accuracy the direction of the antiferromagnetic components is perpendicular to the propagation vector $\boldsymbol{q} = (0, 0, \frac{1}{2})$.

As discussed in ref. [8] such a magnetic diffraction pattern can be explained in terms of three different

[1] Also at Laboratoire Louis Néel, C.N.R.S., 166X, 38042 Grenoble-Cedex, France.

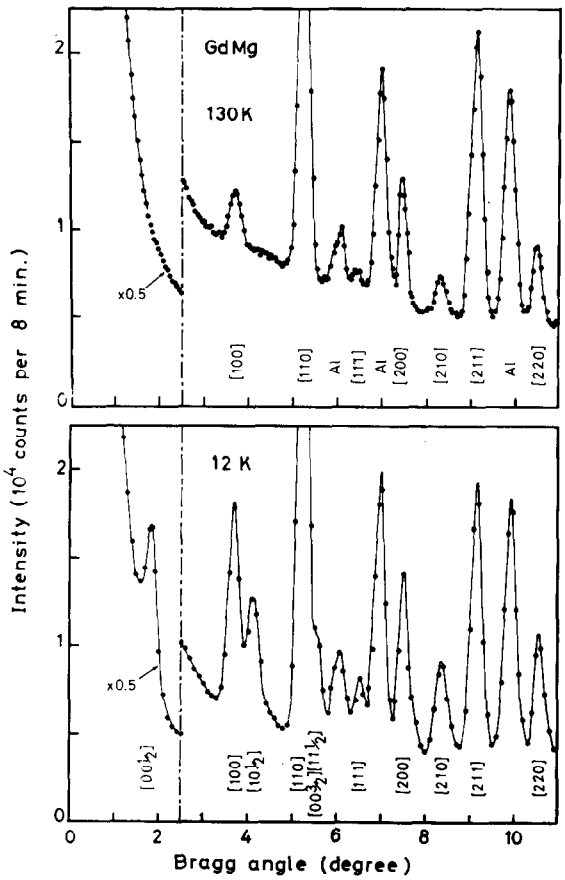

Fig. 1. Neutron diffraction spectra at 12 and 130 K, $\lambda = 0.5$ Å.

types of structure, arising from either a canted structure, or an alternate ferromagnetic and paramagnetic layers arrangement, or an incoherent mixture of ferromagnetic and antiferromagnetic domains. The second type of structure has to be rejected because it leads to a moment larger than the free ion value of 7 μ_B. For the same reason, any single phase structure implies that the μ_F and μ_{AF} components are perpendicular. The dependence of the magnetic intensities (fig. 2) on a magnetic field applied parallel to $\boldsymbol{k}$ [9] allows us to reject the possible mixture of domains and to fully solve the magnetic structure, as described below.

The easy direction for the ferromagnetic component appears to be [111]: the rapid decrease of the (111) magnetic line, in fields up to 400 Oe, is associated with the growing of the domain for which the magnetization is parallel to $\boldsymbol{k}$, at the expense of the seven others. Alternatively the intensity of the (100) magnetic line

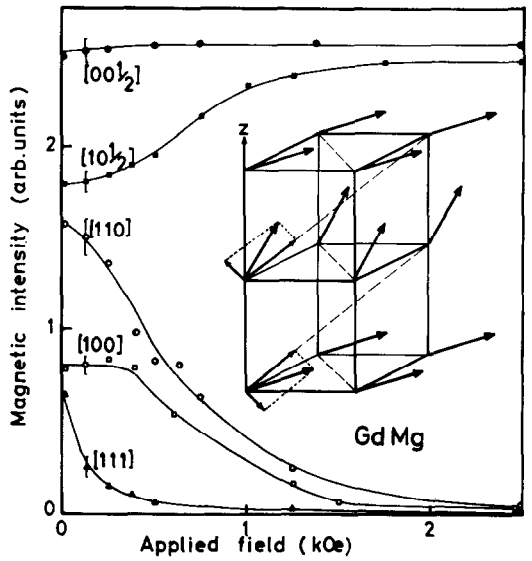

Fig. 2. Field dependence of the different magnetic intensities; a different scale was chosen for each line. Canted magnetic structure of GdMg.

does not vary in low fields since four out of the eight ⟨111⟩ directions are favoured, and the projection of the moments perpendicular to [100] is not changed. In higher fields the intensity of the (100) peak decreases as the ferromagnetic components rotate into the field direction. From the field dependence the anisotropy field is estimated to be 2 kOe. The field dependence of the (110) peak is logically explained in terms of domain wall displacements followed by the rotation of the ferromagnetic components.

The field dependence of the antiferromagnetic lines reveals the perpendicular coupling of the antiferromagnetic and ferromagnetic components. Since the intensity of the $(00\,\frac{1}{2})$ line is field independent, no displacement of antiferromagnetic walls occurs and μ_{AF} is confirmed to be perpendicular to the [001] direction in zero field. On the application of a magnetic field parallel to $(10\,\frac{1}{2})$, the intensity remains constant up to 400 Oe after which it increases and saturates at about 2 kOe. The behaviour is closely related to that observed for the three ferromagnetic lines: above 400 Oe the increase of intensity is explained by the rotation of the antiferromagnetic components driven by that of the ferromagnetic component.

This perpendicular coupling of the μ_F and μ_{AF} components in a single magnetic phase indicates that

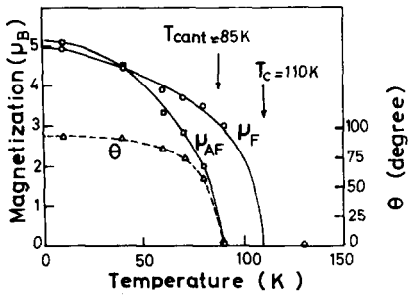

Fig. 3. Thermal dependence of ferromagnetic and antiferromagnetic components and of the angle θ between the moments of the two sublattices.

the magnetic structure of GdMg is canted. As μ_{AF} is perpendicular to the [111] direction and to the $(00\frac{1}{2})$ propagation vector, it must lie along the $[1\bar{1}0]$ direction (inset of fig. 2).

In order to determine the thermal evolution of the magnetic structure, the temperature dependence of the (100) and $(10\frac{1}{2})$ lines was studied. From the results shown in fig. 3, it is clear that μ_{AF} vanishes at $T_{cant} = 85$ K whilst μ_F remains until $T_c = 110$ K. The canted magnetic structure progressively closes when the temperature increases and eventually becomes a collinear ferromagnet between T_{cant} and T_c.

To our knowledge this is the first magnetic structure completely determined on a compound with Gd. A canted structure in an S-ion is somewhat puzzling since in cubic CsCl structure it cannot be explained by the isotropic (Heisenberg) or anisotropic bilinear exchange. In GdMg the coupling responsible for the canted structure is large: at 4.2 K, a field of about 300 kOe is necessary to reach saturation. However, this coupling is weakly anisotropic: from the neutron study, the μ_F component is directed along the field direction for field values larger than 2 kOe, without modification of the canting angle θ. At last, the absence of the canted structure between 85 K and the ferromagnetic ordering temperature suggests a different thermal dependence for the coexisting interactions.

A canted structure has previously been observed in TbMg [3] and in a systematic study of the ternary $TbCu_{1-x}Zn_x$ system [10] similar canted configurations have also been observed, in the region between the antiferromagnetic phase ($x < 0.54$) and the ferromagnetic one ($x > 0.68$). However in these cases CEF effects are strong which makes the analysis more complex.

The canting ordering observed in $TbCu_{1-x}Zn_x$ has been interpreted in terms of the s–f exchange model [11]. In the case of a narrow band, this model reduces to the double-exchange mechanism [12] which has been applied to $Cr_{1-x}Mn_xSb$. However a quite simple explanation may be invoked considering exchange terms of higher order than Heisenberg exchange terms, as a matter of fact a biquadratic coupling [13] is already sufficient to stabilize a canted structure.

References

[1] K.N.R. Taylor, Adv. Phys. 20 (1971) 551.
[2] K.H.J. Buschow and C.J. Schinkel, Solid State Commun. 18 (1976) 609.
[3] R. Aleonard, P. Morin, J. Pierre and D. Schmitt, Solid State Commun. 17 (1975) 599.
[4] R.M. Moon, W.C. Koehler, J.W. Cable and H.R. Child, Phys. Rev. B5 (1972) 997.
[5] J.W. Cable and E.O. Wollan, Phys. Rev. 165 (1968) 733.
[6] Y. Ishikawa, N. Watanabe, K. Tajima and H. Sekine, Phys. Lett. 48A (1974) 159.
[7] A.J. Freeman and J.P. Desclaux, Intern. J. Magn. 3 (1972) 311.
[8] W. Schafer, G. Will and K.H.J. Buschow, J. Magn. and Magn. Mater. 4 (1977) 28.
[9] E.O. Wollan and W.C. Koehler, Phys. Rev. 100 (1955) 545.
[10] T. Yashiro, Y. Hamaguchi and H. Watanabe, J. Phys. Soc. Japan 40 (1976) 63.
[11] M. Umehara and T. Kasuya, J. Phys. Soc. Japan 40 (1976) 13.
[12] P.G. De Gennes, Phys. Rev. 118 (1960) 141.
[13] T. Iwashita and N. Uryu, Phys. Rev. B14 (1976) 3090.