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## Mn N.M.R. AND MAGNETIC STRUCTURES IN $(Y_{1-x}Tb_x)Mn_2$ COMPOUNDS

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**Abstract.** -  $YMn_2$  presents an incommensurate angular modulated magnetic structure while  $Y_{0.9}Tb_{0.1}Mn_2$  is purely antiferromagnetic. The effect of the modulation on the anisotropic hyperfine field leads to a large and asymmetric Mn N.M.R. spectrum on  $YMn_2$  while a sharper and symmetric one is observed in  $Y_{0.9}Tb_{0.1}Mn_2$ .

$YMn_2$  crystallizes in the FCC Laves phase (Fd3m). Thermal expansion [1] and magnetization [2] measurements show that  $YMn_2$  has a first order antiferromagnetic transition around 100 K with a large volume anomaly (5 %). In a previous paper [3] we have discussed the magnetic structure deduced from neutron diffraction, associated with results obtained by N.M.R. [4]. Below  $T_N$  the neutron diffraction patterns revealed diffraction reflections which are characteristic of the antiferromagnetic ordering of the Mn moments. The center of gravity of all the observed magnetic peaks can be indexed with  $h+k$  or  $k+l = 2n+1$ ; furthermore a precise analysis indicates that they are splitted in two or several peaks (Fig. 1a). The deduced magnetic structure is helimagnetic type, incommensurate, with a very long period ( $\sim 200 \text{ \AA}$ ). The N.M.R. spectrum obtained at 4.2 K by Yoshimura *et al.* [4] presents two peaks at 120 and 130 MHz with different intensities. We have explained it [3] as arising from an anisotropy of the hyperfine field due to an anisotropy of the Mn magnetization and from a perturbation of the helix by the magnetocrystalline anisotropy.

Recently we have studied the properties of the pseudobinary  $(Y_{1-x}Tb_x)Mn_2$  compounds [5]. For  $x = 0.1$

the neutron diffraction pattern observed at low temperature is very similar to that of pure  $YMn_2$ . However the splitting of the magnetic peaks is no more present, thus the magnetic structure is purely antiferromagnetic. The possible arrangements of the Mn moments, deduced from the refinement of the measured intensities [5] are similar to the local magnetic arrangements in pure  $YMn_2$ ; but the angular modulation which is associated to the helix, is not observed in  $Y_{0.9}Tb_{0.1}Mn_2$ . In this paper we discuss the effect of these different magnetic structures on the shape of the N.M.R. spectra in  $YMn_2$  and  $Y_{0.9}Tb_{0.1}Mn_2$ .

Polycrystalline samples have been prepared by induction melting in a water-cooled crucible followed by 3 days of annealing at 850 °C. The zero field N.M.R. study of <sup>55</sup>Mn was performed at 1.4 K using a pulsed spin echo spectrometer in the frequency range from 100 to 200 MHz. Spin echoes were excited with rather long pulses ( $\Pi/2 = 8 \mu s$ ) and high radiofrequency (R.F.) power (300 W). These conditions are characteristics of a signal in antiferromagnetic (A.F.) compound with low R.F. enhancement factor.

In the spectrum obtained in the pure  $YMn_2$  (Fig. 2a), the shape of the resonance line centered at 120 MHz is similar to that observed by Yoshimura and Nakamura [4], while instead of a second peak at 130 MHz, only a long asymmetric tail is found. In order to clarify the origin of extra line at 130 MHz detected by [4] we extended our measurements on compounds whose compositions present a defect ( $YMn_{2-\epsilon}$ ) or an excess ( $YMn_{2+\epsilon}$ ) of manganese. No significant change occurs in the compound which is deficient in Mn, while a strong and narrow signal, superposed on the wide A.F. echo, appears in the sample rich in Mn. The superposed echo was optimized with short pulses ( $\Pi/2 = 0.5 \mu s$ ) and low R.F. power which indicates that it is of ferromagnetic origin. We attributed the signal to  $Y_6Mn_{23}$  phase, referring to [4] where this ferromagnetic phase gave the resonance lines at 90, 112

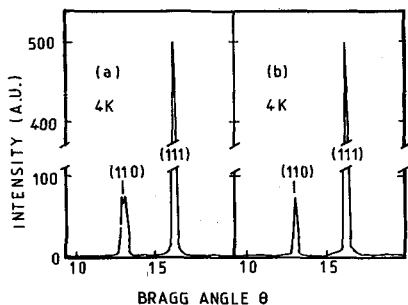


Fig. 1. - Neutron diffraction: detail of the [110] magnetic line compared to the [111] nuclear one: a) in pure  $YMn_2$  and b) in  $Y_{0.9}Tb_{0.1}Mn_2$ .

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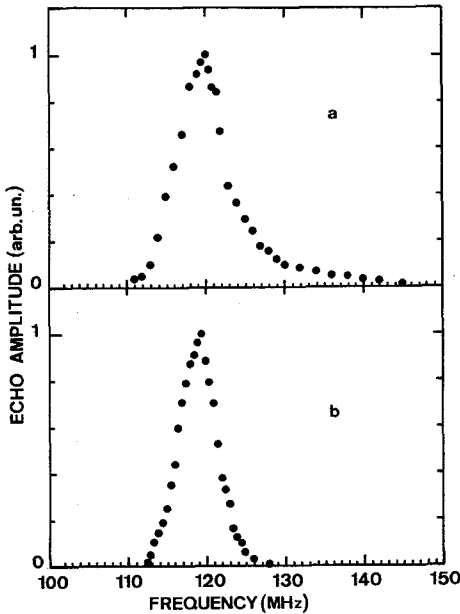


Fig. 2. - Zero field N.M.R. spectra of  $^{55}\text{Mn}$  a) in pure  $\text{YMn}_2$  and b) in  $\text{Y}_{0.9}\text{Tb}_{0.1}\text{Mn}_2$  at 1.4 K. The spin echo amplitude is corrected for the  $\nu^2$  frequency dependence.

and 130 MHz. We also suspect that some traces of the  $\text{Y}_6\text{Mn}_{23}$  phase in the  $\text{YMn}_2$  sample investigated in [4] might have induced the observation of peak at 130 MHz.

Returning to the analysis of the asymmetric spectrum of  $\text{YMn}_2$  (Fig. 2a), we note that an A.F. helical spin structure (AFHS) can be analysed assuming a planar anisotropy of the hyperfine field:

$$H_{\text{hf}}(\theta) = H_{\text{hf}}(1 - p \sin^2 \theta) \quad (1)$$

where  $\theta$  is the angle between the direction of the easy magnetization and the direction of magnetic moments and  $p$  the hyperfine field anisotropy which result from the anisotropy of the Mn magnetization.

The broadening due to the anisotropic quadrupolar splitting:

$$\delta V_Q = V_Q(3 \cos^2 \theta - 1) / 2 \quad (2)$$

is expected to be negligible.

A simulation of the spectrum assuming a Gaussian shape for the individual resonance lines and an isotropic distribution of angles  $\theta$  (in the plane) gives two peaks corresponding to  $\theta = 0$  and  $\theta = \Pi/2$ . Such a spectrum was actually observed in  $\text{MnSi}$  [6]. In the spectrum of  $\text{YMn}_2$  (Fig. 2a) instead of a second peak which would correspond to  $\theta = \Pi/2$ , along a long tail is observed. It is worthy to note that in the tail the transverse relaxation time  $T_2$  was found to decrease, indicating that these frequencies correspond to angles  $\theta \neq 0$  in the above mentioned model. The observed shape of the spectrum corresponds to the magnetic moments essentially distributed inside an angle  $\theta < \theta_{\text{max}}$  around the easy magnetization direction for each of the A.F. sublattices. The fact that  $\theta$  does not take all values from 0 to  $\Pi/2$  is due to the magnetocrystalline anisotropy of the Mn atoms which perturbs the regular helimagnetic arrangement. This picture is supported by the changes in the spectrum induced by the substitution of 10 % of Y by Tb. In the N.M.R. spectrum of  $\text{Y}_{0.9}\text{Tb}_{0.1}\text{Mn}_2$  shown in figure 2b (compared to Fig. 2a) the  $^{55}\text{Mn}$  line is now centered on 119.5 MHz and becomes symmetric and sharper, in agreement with the pure A.F. structure deduced by neutron diffraction.

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