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To cite this version:

R. Chakravarthy, L. Madhav Rao, S. Paranjpe, S. Kulshreshta, A. Soper, et al.. MAGNETIC STRUCTURE OF $\text{Co}_{0.5}\text{Zn}_{0.5}\text{FeCrO}_4$. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1111-C8-1112. <10.1051/jphyscol:19888510>. <jpa-00228717>

HAL Id: jpa-00228717

https://hal.archives-ouvertes.fr/jpa-00228717

Submitted on 1 Jan 1988

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MAGNETIC STRUCTURE OF Co$_{0.5}$Zn$_{0.5}$FeCrO$_4$

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Abstract. — Neutron diffraction studies on the disordered spinel Co$_{0.5}$Zn$_{0.5}$FeCrO$_4$ over a temperature range 320 K to 20 K show this system to be ferrimagnetically ordered, passing on to a semi spin glass state around 20 K.

1. Introduction

Recently, several disordered concentrated magnetic systems have been shown to exhibit spin glass behaviour [1]. Among these, the spinel structure systems are more interesting since they offer the possibility of selective magnetic dilution of the tetrahedral (A) and octahedral (B) sites which can lead to a large variety of magnetic behaviour. Although a systematic neutron diffraction study of spin glass spinels with magnetic ions occupying only the octahedral sublattice has been reported [2], similar studies with magnetic ions on both the sublattices are scarce. To understand the magnetic structure of one such spinel we have carried out neutron diffraction studies on Zno.$\cdot$5FeCrO$_4$ from 320 K down to 20 K. This composition was selected for the following reason: the A site magnetic ion concentration is more than the A sublattice percolation threshold of 0.429 [3] and hence one expects long range ferrimagnetic ordering in this system. However, ac susceptibility, low field dc magnetisation and Mössbauer studies have shown that this system has no long-range magnetic ordering but transforms into a cluster spin glass state below 140 K [4].

2. Experimental

The sample was prepared using the wet chemical method, and the X-ray diffraction lines showed a well formed homogeneous spinel phase. AC susceptibility measurement done on a portion of the sample showed a broad distribution with the peak around 140 K, with the signal being non zero even at 25 K. These features agree broadly with the earlier measurement [4]. Neutron diffraction measurements were made on the Liquid and Amorphous Diffractometer (LAD) at the pulsed neutron source at RAL (U.K.) at eleven different temperatures, 320 K to 20 K.

3. Results and discussion

The data taken at 320 K, limited to higher $Q$ region (where magnetic contributions are quite negligible) was profile analysed using the Rietveld technique to arrive at the following cation distribution: (Zn$_{0.5}$Co$_{0.35}$Fe$_{0.12}$) on the A site and (Co$_{0.07}$Fe$_{0.47}$Cr$_{0.5}$)$_2$ on the B site. The weighted profile $R$ factor is 5 %. The inner Bragg peaks were seen to have significant magnetic contribution (at temperatures below 320 K) and their widths were within the instrumental resolution indicating clearly the existence of long range ferrimagnetic ordering. The Bragg intensities of the patterns, at the different temperatures were least squares analysed and the magnetic structure factors of the relevant reflections extracted. At 20 K, the A site moment is 2.2(2) $\mu_B$ indicating a significant orbital contribution (due to the Co$^{3+}$ ion) since the estimated spin-only moment is 1.7 $\mu_B$. The

Fig. 1. — Temperature dependence of the magnetic structure factor for the (220) reflection (A site) and the (222) reflection (B site). Continuous curves are the Brillouin functions.
B site moment at the same temperature is 1.7(2) \( \mu_B \), substantially smaller than the estimated “spin-only” moment of 3.8 \( \mu_B \). The magnetic structure is therefore one where the A site moments are all collinear whereas the B site moments are highly non collinear. The site moments deduced at all other temperatures are consistent with this magnetic configuration, \( T_N \) being around 320 K. Figure 1 shows the “reduced” magnetic structure factor (magnetic structure factor at any given temperature divided by that at 20 K) versus the “reduced” temperature for the (220) reflection (purely A site contribution) and for the (222) reflection (purely B site contribution). It is observed that the temperature dependence of these site moments do not follow the smooth trend that one expects for a “classical” ferrimagnetic ordering, but is characterized by steps indicating a sluggish temperature response of the sublattice magnetisation at these temperatures. The absence of a sharp (200) magnetic reflection at \( Q = 1.5 \text{ Å}^{-1} \) indicates that the transverse components of the B site moments are disordered. However, as seen in figure 2a broad diffuse hump appears at this region as one approaches 20 K reflecting a build-up of short range correlation of these transverse components around 20 K.

The present neutron study reveals that from \( T_N \) down to 20 K, \( \text{Zn}_{0.5}\text{Co}_{0.5}\text{FeCrO}_4 \) is ferrimagnetic with non-collinear B site moments, its transverse components being randomly oriented. Around 20 K freezing of these components occur. This situation which involves longitudinal long range ferrimagnetic ordering coupled with random freezing of the transverse components of the magnetic moments seems to resemble the semi-spin glass state as discussed by Villain [5]. Neutron measurements below 20 K are in progress and will be reported elsewhere.

Fig. 2. – Difference intensity patterns for three temperatures showing the build up of the diffuse hump at 20 K.