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## MAGNETISM OF RANDOMLY CANTED Li-Ti FERRITE

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**Abstract.** – FC-ZFC magnetisation, hysteresis, thermoremanence and neutron diffraction measurements were performed on  $\text{Li}_{1.125}\text{Fe}_{0.625}\text{Ti}_{1.25}\text{O}_4$ . The results are interpreted from domain and wall properties resulting of local canted state where the spin transverse component relaxes between preferential directions.

### Introduction

The dilution problem in substituted two magnetic sublattice system has attracted considerable interest. A local canted state (LCS) was first presented by Rosencwaig [1] and refined by Patton [2]. Villain's model [3] predicted a LCS embedded within a collinear ferrimagnetic lattice. This state is expected to undergo two transitions: one at  $T_N$  for the longitudinal spin component  $S_z$  (collinear order) and a second one at  $T_g$  for the transverse component  $S_t$  (spin glass type order). Recently two transitions have been reported for the systems  $\text{Zn}_x\text{Mg}_{1-x}\text{Fe}_2\text{O}_4$  [4] and  $\text{Mg}_{1+t}\text{Fe}_{2-2t}\text{Ti}_t\text{O}_4$  [5].

Our study on the system  $\text{Li}_{0.5+0.5t}\text{Fe}_{2.5-1.5t}\text{Ti}_t\text{O}_4$  [6] has shown that, for  $t = 1.25$ , both Fe spins at A and B sites are canted. High magnetisation and Mössbauer results are interpreted by a thermally activated relaxation of  $S_t$  between preferential directions.  $S_t$  is not free to rotate; this deviates from the Villain's model. We present FC-ZFC static magnetisation, hysteresis, thermoremanence and neutron diffraction measurements.

### Results and discussion

The neutron diffraction experiment was performed between 300 and 4.2 K. The nuclear spectra ( $R \sim 1.5\%$ ) give an atomic distribution in agreement with the X-ray and Mössbauer results. Below  $T_N \approx 110$  K, unfortunately only a very weak magnetic contribution is superimposed to the (111) peak. This can be qualitatively related to a  $S_z$  ferrimagnetic order. FC-ZFC magnetisation measurements were performed with  $3.4 \text{ Oe} \leq H \leq 138 \text{ Oe}$ , and hysteresis loops with  $H \leq 300 \text{ Oe}$  and  $4.2 \text{ K} \leq T \leq 110 \text{ K}$ . Figure 1 shows that the FC-ZFC branching is strongly field dependent: for  $H < 30 \text{ Oe}$ , irreversibilities are observed until  $T_N$ ; for  $H \geq 30 \text{ Oe}$ , FC-ZFC values are identical above the magnetisation maximum temperature  $T_{\text{max}}$ . This behavior cannot be related to (i) a spin glass regime because the sample is a perturbed ferrimagnet, (ii) a magnetic cluster model because the branching point is

too sensitive to  $H$ . It can be rather a consequence of the hysteresis loop characteristics which depend on  $T$ . For a given  $T$  and  $H$ , if the field-magnetisation ( $M-H$ ) curves are irreversible, FC and ZFC differ; if  $M-H$  are reversible, they can be equal. The branching point depends on the hysteresis loop evolution which is related

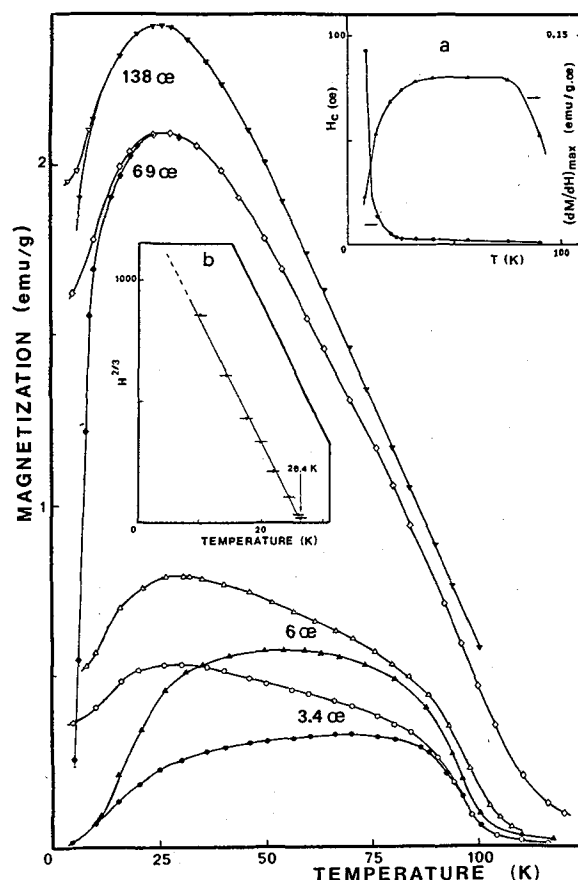


Fig. 1. –  $T$  and  $H$  dependence of the FC-ZFC magnetisation. Inserts; (a)  $T$  dependence of the coercive field  $H_c$  and the maximum slope  $(dM/dH)_{\text{max}}$  of the hysteresis loop. (b)  $H$  dependence of the FC magnetisation maximum temperature.

to the magnetic domains. But for  $T \leq 20$  K, the ZFC and  $(dM/dH)_{\max}$  drops, like in reentrant systems, can be related to a disorder phenomenon, although they are time dependent. These features are discussed below.

The FC maximum temperature depends on  $H$  :  $T_{\max} = T_0 - (1.97 \times 10^{-2}) H^{2/3}$ , with  $T_0 = 26.4$  K and  $50 \text{ Oe} \leq H \leq 25 \text{ kOe}$ . This behavior is found in spin glasses and in small particles, but could also be more general if the maximum is related to an activated phenomenon as the  $S_t$  relaxation.

FC values are thermic hystory dependent, just below  $T_N$ , in particular when the measurements are performed in decreasing  $T$  with different cooling rates. Considering that  $S_t$  has preferential positions [6], the system energy has probably several minima depending on  $T$ ,  $H$ , cooling rate and measuring method. A magnetisation relaxation, toward a more stable state, can exist but is certainly slow.

For  $T > T_{\max}$  and  $H \geq 20$  Oe, the hysteresis loop is closed and the FC-ZFC curves show (i) a magnetisation increase with  $H$  partly due to a weak canting angle variation, (ii) a magnetisation decrease when  $T$  increases which cannot be easily explained because the two sublattice existence and a possible canting variation with  $T$ . The presence of some  $S_z$  population, with a fast relaxation, cannot be exclude, this population decreasing with  $T$ .

The time dependence of the thermoremanence magnetisation has been studied for applied fields  $H = 3.4, 28, 69$  and  $138$  Oe with  $4.2 \leq T \leq 90$  K. The cooling time, from  $300$  K to  $T$ , is about  $25$  mn, and the measuring time  $\sim 10^3$  s. The phenomenological relation is observed:  $\text{TRM}(T, t) = C(T) \exp[-p(T) \ln t]$ .  $p(T)$  increases with  $T$  as in spin glass regime, then decreases above  $T \sim 20$  K which is not usually observed. To explain the  $\text{TRM}(T, t, H)$  behavior, we have used the Khater relation [7] giving  $\text{TRM}(T, t)$  from a cluster model, which permitted to determine the mean activation energy  $E_a/k$ . The  $T$  dependence of  $E_a/k$  is presented in figure 2. For  $T \leq 15$  K,  $E_a/k$  is constant:  $(E_a/k)_0 = 280$  K. The high temperature behavior can be interpreted by adding to  $(E_a/k)_0$  a supplementary term inversely proportional to the relaxation time  $\tau_t$  of  $S_t$ . The disorder introduced by  $S_t$  could explain the low temperature-TRM behavior, while at high  $T$  the relaxation is fast and the order evolves toward a classical ferrimagnet.

All reported features can be explained from the domain and wall properties resulting of LCS. At low  $T$ ,  $S_t$

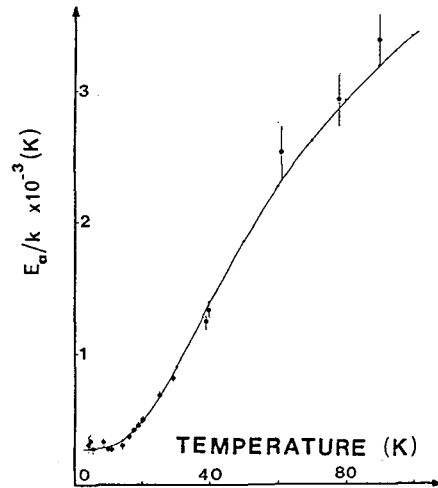


Fig. 2. -  $T$  dependence of the activation energy  $E_a/k$ .

is frozen in an energy minimum direction and it is difficult that the walls move. Indeed, after spin rotation,  $S_t$  is not again in a direction minimizing the energy and therefore the energy cost is high. Then  $H_c$  is high and  $(dM/dH)_{\max}$  low. Nevertheless the system relaxes and an equilibrium state is reached after a certain time, then a time variation of  $H_c$  and  $(dM/dH)_{\max}$  is observed and a decay of TRM. At high  $T$ , the energy barrier between the preferential directions is low, the  $S_t$  relaxation is fast. Therefore the equilibrium state is rapidly reached and the properties are similar to those of a classical ferrimagnet.

In conclusion, all the particular properties of this compound result of the existence of LCS where the transverse component relaxes between preferential directions.

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