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## FIRST ORDER MAGNETIZATION PROCESS IN $\text{Sm}(\text{Fe}_{11}\text{Ti})$

Hong-Shuo Li <sup>(1)</sup>, Bo-Ping Hu <sup>(1)</sup>, J. P. Gavigan <sup>(1)</sup>, J. M. D. Coey <sup>(1)</sup>, L. Pareti <sup>(2)</sup> and O. Moze <sup>(2)</sup>

<sup>(1)</sup> Department of Pure and Applied Physics, Trinity College, Dublin 2, Ireland

<sup>(2)</sup> MASPEC Institute of C.N.R., 43100 Parma, Italy

**Abstract.** – Magnetization measurements on the  $\text{ThMn}_{12}$ -structure compound,  $\text{Sm}(\text{Fe}_{11}\text{Ti})$ , reveal a sharp transition below 150 K when a field of 9 T is applied perpendicular to the *c*-axis. Singular point detection data indicate a type-II first-order magnetization process, which is incompatible with a pure  $^6\text{H}_{5/2}$   $\text{Sm}^{3+}$  ground state. A model which takes account of mixing of higher *J*-multiplets is used to explain the observations.

The new tetragonal  $\text{ThMn}_{12}$ -structure compound  $\text{Sm}(\text{Fe}_{11}\text{Ti})$  has recently aroused great interest. The Curie temperature of 584 K [1] is nearly the same as that of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , 589 K. Its strong uniaxial anisotropy and high iron-content make  $\text{Sm}(\text{Fe}_{11}\text{Ti})$  potentially suitable for permanent magnets. Here we report that the compound exhibits a discontinuous change in magnetization when a magnetic field is applied in a hard direction at low temperature. This first order magnetization process (FOMP) has interesting implications for the electronic state of samarium in the compound.

Oriented samples of  $\text{Sm}(\text{Fe}_{11}\text{Ti})$  were made by mixing finely-ground powder with epoxy resin and setting in a field of 1.5 T. Magnetization curves were measured with the applied field perpendicular to the orientation direction in fields up to 15 T, at the High Field Magnet Laboratory of University of Nijmegen and at the Service National des Champs Intenses, Grenoble. Measurements in pulsed fields using the singular point detection technique (SPD) were carried out in Parma. Some of the magnetization curves obtained as a func-

tion of temperature are shown in figure 1. A sharp upturn is seen in an applied field of about 9 T below 150 K. The SPD measurements indicate the FOMP is of type II (i.e. the magnetization is not saturated after the transition, like that of  $\text{Pr}_2\text{Fe}_{14}\text{B}$  [2]). The thermal variation of the critical and anisotropy fields deduced from these measurements are shown in figure 2.

Previous magnetic measurements on  $\text{Y}(\text{Fe}_{11}\text{Ti})$  [1, 3] have shown that the anisotropy of the iron sublattices favours the *c*-axis ( $K_1^{\text{Fe}} > 0$ );  $^{155}\text{Gd}$  Mössbauer results on  $\text{Gd}(\text{Fe}_{10}\text{T}_2)$  [4] and a spin reorientation study of  $\text{Dy}(\text{Fe}_{11}\text{Ti})$  [5] suggests that  $A_2^0$  for the 2a site in  $\text{ThMn}_{12}$  structure is negative, (opposite to that at rare earth sites in the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  structure). Hence the second order crystal-field for  $\text{Sm}^{3+}$  also favours the *c*-axis ( $K_1^{\text{R}} > 0$ ). The total second-order anisotropy in zero field,  $K_1 = K_1^{\text{Fe}} + K_1^{\text{R}}$ , should therefore be positive in  $\text{Sm}(\text{Fe}_{11}\text{Ti})$ . It was confirmed by Mössbauer spectroscopy on an oriented sample that a *c*-axis orientation of better than 90 % was achieved. Following the theory developed by Asti and Bolzoni [6], the type II FOMP can be understood using the phenomenological anisotropy constants  $K_1, K_2, K_3$ . For a system with positive  $K_1$ , the type II FOMP requires negative  $K_2$  and positive  $K_3$ . Fitting the critical field and the

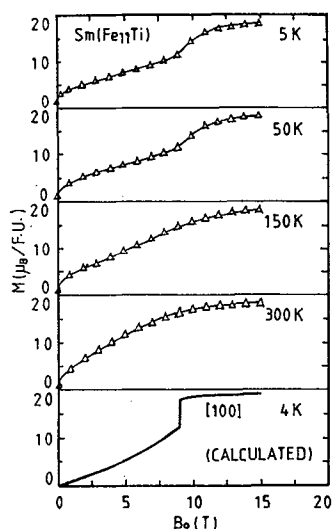


Fig. 1. – High field magnetization curves of  $\text{Sm}(\text{Fe}_{11}\text{Ti})$  at different temperatures.

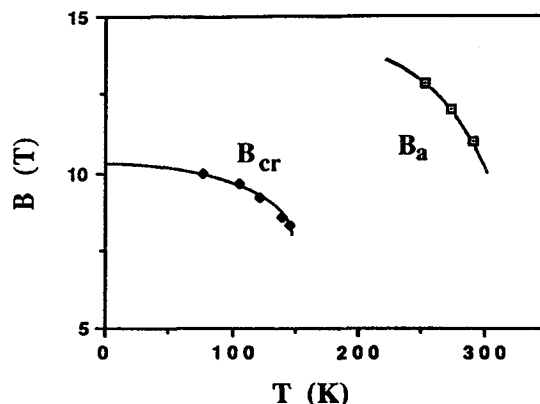


Fig. 2. – Thermal variation of the experimental anisotropy and critical fields in  $\text{Sm}(\text{Fe}_{11}\text{Ti})$ .

jump in magnetization we find  $K_1 = 160$  K,  $K_2 = -96$  K and  $K_3 = 34$  K at 4.2 K. However  $K_3^R$ , which is related to the sixth-order crystal field, is zero for the  ${}^6\text{H}_{5/2}$  ground state multiplet of the  $\text{Sm}^{3+}$  ion (the sixth order Stevens coefficient  $\gamma_J = 0$  for  $J = 5/2$ ).

We have established an exchange and crystal-field model to account for available experimental data on the R( $\text{Fe}_{11}\text{Ti}$ ) series. The first excited multiplet  ${}^6\text{H}_{7/2}$  of the  $\text{Sm}^{3+}$  ion is only 1438 K [7] above the ground state  ${}^6\text{H}_{5/2}$ . It is therefore necessary to take account of  $J$ -mixing as in the case of  $\text{SmM}_5$  ( $M = \text{Co}$  [8, 9],  $\text{Ni}$  [10]). The total Hamiltonian for the  $\text{Sm}^{3+}$  ion can be written as

$$H = \lambda \mathbf{L} \cdot \mathbf{S} - 2 \mu_B \mathbf{S} \cdot \mathbf{n}_{\text{SmFe}} \langle M_{\text{Fe}} \rangle + H_{cf} - \mu_B (\mathbf{L} + 2\mathbf{S}) \cdot \mathbf{B}_0 \quad (1)$$

The first term above is due to spin-orbit coupling with  $\lambda = 411.1$  K [7]; the second term represents the exchange interaction between Sm and Fe sublattices, with an exchange coefficient  $\mathbf{n}_{\text{SmFe}}$  deduced from the Curie temperature [11] of 300  $\mu_0$ ; the third term is the crystal-field Hamiltonian for the 2a site in the tetragonal  $\text{ThMn}_{12}$ -structure having point symmetry  $4/\text{mmm}$

$$H_{cf} = N_2^0 A_2^0 \langle r^2 \rangle U_0^2 + N_4^0 A_4^0 \langle r^4 \rangle U_0^4 + N_4^4 A_4^4 \langle r^4 \rangle U_4^4 + N_6^0 A_6^0 \langle r^6 \rangle U_0^6 + N_6^4 A_6^4 \langle r^6 \rangle U_4^6 \quad (2)$$

Here  $\{A_k^q\}$  are the crystal-field coefficients,  $\langle r^k \rangle$  is an average over the 4f orbitals given by Freeman and Desclaux [12], and the Racah operators  $\{U_q^k\}$  are deduced using 3-j and 6-j symbols [13]. The fourth term in equation (1) is the Zeeman term where  $\mathbf{B}_0$  is applied field. Our method of calculation is the same as that used for  $\text{Nd}_2\text{Fe}_{14}\text{B}$  [14]. The iron sublattice magnetization ( $M_{\text{Fe}}$ ) is taken from that of  $\text{Y}(\text{Fe}_{11}\text{Ti})$  [1] and the ratios of  $A_4^4/A_4^0$  and  $A_6^4/A_6^0$  are fixed from point charge calculation as  $-2.4$  and  $1.6$  respectively. The FOMP can be explained with the set of  $\{A_k^q\}$  which is listed in table I. These values are all larger than those reported previously for  $\text{Dy}(\text{Fe}_{11}\text{Ti})$  [5]. The corresponding calculated magnetization curve at 4.2 K for applied field along [100] is presented in figure 1.

In fact,  $J$ -mixing leads to a non-collinear structure for the magnetic moment  $\mathbf{M} = (\mathbf{L} + 2\mathbf{S})$  and spin  $\mathbf{S}$ .

Table I. - Values of  $\{A_k^q\}$  obtained to describe FOMP, in  $K \cdot a_0^{-k}$

$A_2^0$	$A_4^0$	$A_6^0$
-170	27	-16

Keeping in mind that the exchange interaction always couples the spins while Zeeman interaction acts on  $\mathbf{M}$ , the non-collinearity may become large when an applied field is present, resulting in large, field-dependent values of  $\{K_i\}$  [15]. We can conclude that large positive  $K_3$  required for FOMP comes not only from sixth-order crystal-field but also from competition between the exchange and Zeeman interactions.

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