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LETTER TO THE EDITOR

Anisotropy and magnetic ordering in the new phase $Nd_3(FeTi)_{29}$

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Abstract. Recently, a new rare-earth iron-rich phase 3:29 has been found to be stabilized using Ti. The Curie temperature $T_C = 413$ K is considerably higher than in the related Nd_2Fe_{17} compound. The easy magnetization direction lies in the basal plane of the monoclinic structure at room temperature.

A magnetic anomaly has been detected at $T_{SR} = 233$ K in the thermal dependence of the AC initial susceptibility, which has been related to a spin reorientation transition from an easy-plane phase towards an easy cone at low temperatures. Thermal expansion measurements clearly show an extra magnetic contribution at T_C, a weak anomaly being also perceptible at T_{SR}. The anisotropy field measurements using the SPD technique have confirmed the existence of these two magnetic phases.

Rare-earth iron-rich intermetallics are being widely investigated since the discovery of the $R_2Fe_{14}B$ phase, which turned the research for new hard magnetic intermetallics towards the stabilization of new iron-rich ternary phases like $R_2(Fe\text{–}M)_{17}$, $R(Fe\text{–}M)_{12}$ etc. The possibility of increasing the intensity of the exchange interaction and also the anisotropy by introducing interstitial atoms has created a great deal of expectation for technical applications of these compounds. In the frame of research for new iron-rich intermetallics, the phase $R_3(FeTi)_{29}$ was discovered [1]. This new phase was initially considered to be $R_2(FeTi)_{19}$, being characterized by x-ray, magnetization and Mössbauer spectroscopy [2, 3]. In a recent work [4] this phase has been identified to be $R_3(FeTi)_{29}$, crystallizing in the monoclinic spatial group $P2_1/C$, which has been also suggested in [5].

In this letter we report AC initial magnetic susceptibility, thermal expansion and anisotropy field measurements on the compound $Nd_3(FeTi)_{29}$.

The polycrystalline sample was prepared from high-purity Nd (3N) and Fe, Ti (4N) using an argon-arc furnace. Further heat treatment is required in order to stabilize the desired phase and we followed the procedure given in the original work of Collocot *et al* [1]. The sample was annealed at 1100 °C in an argon atmosphere for three days and was then water quenched. X-ray diffraction analysis showed the same pattern as that proposed for the 3:29 compounds, with the additional presence of a small amount of α-Fe phase which was also detected by thermomagnetic analysis. Scanning electron microscopy and energy dispersion x-ray microanalysis confirmed that the majority phase had a composition very close to the nominal starting phase, 9.4:86.5:4.1.

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Thermal expansion measurements were performed using a 'push-rod' method in the temperature range 150–550 K. The AC initial magnetic susceptibility was measured using a mutual inductance Harsthorn bridge with an excitation field of approximately 30 mOe of peak value at a frequency of 15 Hz. SPD (singular-point detection) measurements were carried out using a high pulsed magnetic field up to 35 T and temperatures in the range 77–300 K.

Linear thermal expansion (LTE) results are presented in figure 1, where a significant magnetic contribution to the phonon anharmonic Grüneisen contribution can be observed. This contribution is originated by the appearance of magnetic ordering below $T_C = 413$ K. We do not expect any change of the local magnetic moment of the different magnetic sublattices at T_C in this alloy, and, as a consequence, the large magnetovolume effect observed must originate from the dependence of the exchange interaction on volume. The magnitude of the observed spontaneous magnetovolume effect is a guide for the sensitivity of the exchange integrals (J_{Fe-Fe}) to volume. The allocation of interstitial atoms could lead to an increase of volume, producing large effects on the J_{Fe-Fe}, and hence a large increase of the T_C would be expected. In fact, it has been observed that an increase of 5% in volume by nitrogenation results in an increase in T_C of 200 K [1,2].

An additional weak anomaly is observed at 233 K (see figure 1). This barely observed anomaly corresponds to a spin reorientation process in which the easy magnetization direction rotates from the easy plane at room temperature to an intermediate direction between the plane and the c-axis of the crystallographic structure. The existence of this SRT was more evident from AC initial magnetic susceptibility (χ_{AC}) measurements. Figure 2 shows the thermal dependence of χ_{AC}. A clear and distinct peak anomaly is observed at 233 K in close agreement with LTE results. In the inset of figure 2 we display the SRT region in more detail. The SRT is from easy plane to easy cone as determined from SPD measurements and as we will report. Below T_{SR} and around 150 K (see figure 2), a broad shoulder is observed in the thermal dependence of χ_{AC}. SPD measurements have revealed the relation of this anomaly to changes in the anisotropic behaviour.

In figure 3 we display the thermal dependence of the anisotropy fields obtained from SPD measurements. At room temperature the sample is easy plane ($K_1 < 0$) and the anisotropy field $H_A = 2K_1/M_s = 2.8$ T. (M_s is the saturation magnetization and K_1 the phenomenological anisotropy constant). Between room temperature and 233 K only one anisotropy field is observed. At this temperature (T_{SR}) the relation $K_2/K_1 < -0.5$ should be satisfied (with $K_1 < 0$ and $K_2 > 0$). From room temperature down to 200 K the value of K_1 slightly increases, as is evident from the increase of H_A. The occurrence of a SRT is associated with the increase of K_2 being more rapid than that of K_1 for decreasing temperature. Below T_{SR} a second peak is observed in the second derivative of the SPD signal at a magnetic field value H'_A lower than the anisotropy field. This result is consistent with a magnetic state in which the easy magnetization direction is not along a major symmetry direction. In this situation a complex structure exists. The overall easy magnetization direction along an intermediate direction between the c-axis and the basal plane is called the easy cone and is characterized by the existence of two anisotropy fields H_A and H'_A, H_A being the field needed to saturate the sample along the c-axis, and $H'_A = 2(K_1 + 2K_2 + 3K_3)/M_s$ being the field needed to saturate the sample along a direction in the basal plane. H'_A is found to increase linearly with decreasing temperature. As expected, the extrapolated value of the H'_A thermal dependence vanishes at T_{SR} (see figure 3).

Below 100 K, the approach to saturation towards the basal plane develops into a $P1C$-type (saturation along a direction in the basal plane) first-order magnetization process (FOMP).

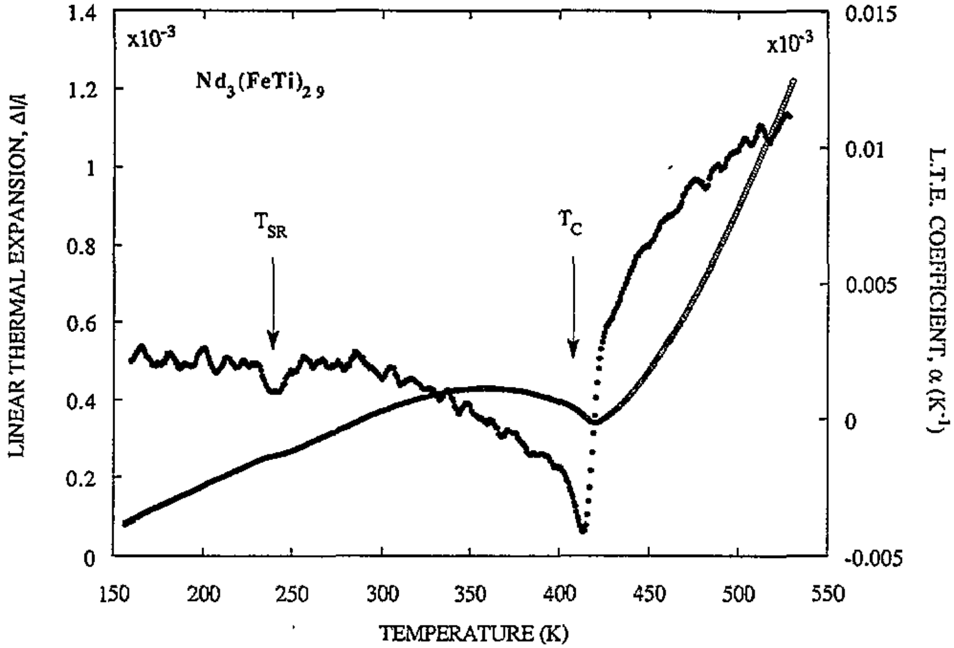


Figure 1. The linear thermal expansion (LTE) and the LTE coefficient of Nd$_3$(FeTi)$_{29}$.

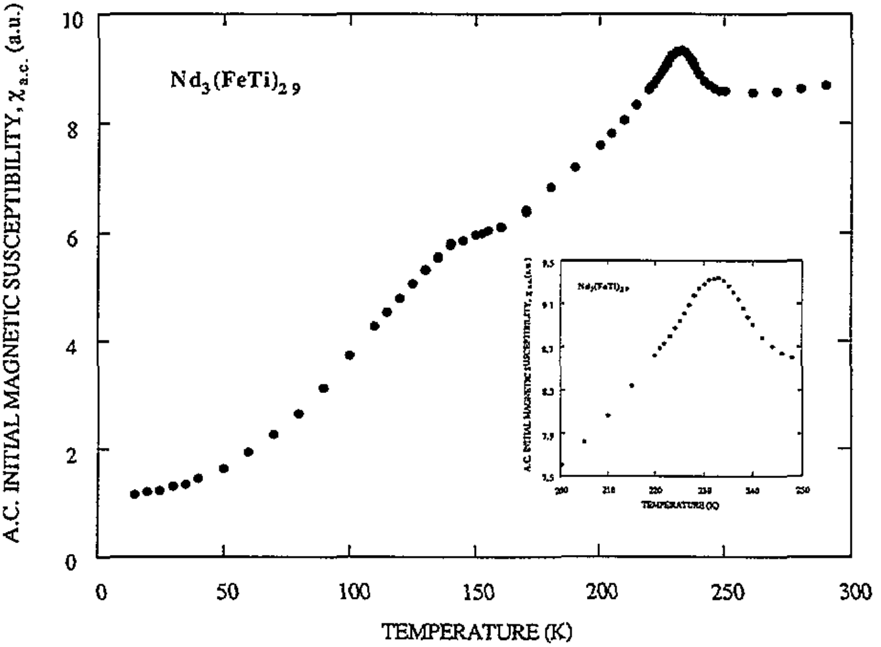


Figure 2. The thermal dependence of the AC initial magnetic susceptibility in Nd$_3$(FeTi)$_{29}$.

Quite recently, such a process has been observed in a magnetically aligned powder sample of

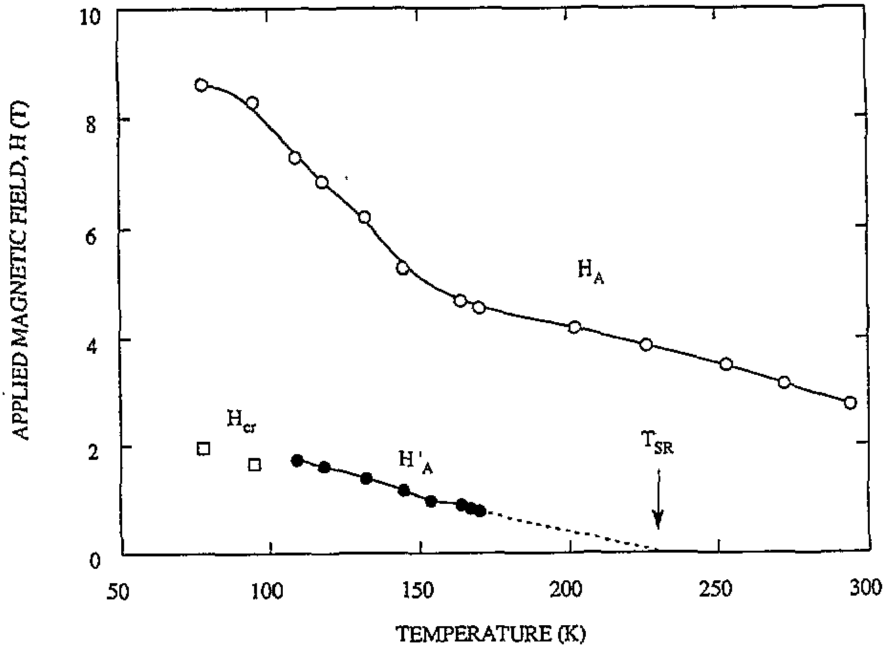
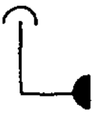


Figure 3. The thermal dependence of the anisotropy fields $H_A = 2K_1/M_s$ and $H'_A = 2(K_1 + 2K_2 + 3K_3)/M_s$ obtained from SPD measurements on $Nd_3(FeTi)_{29}$ (the values reported are in terms of applied magnetic field; the internal field can be calculated using the expression $H^i_A = H_A - 4\pi M_s/3$).

a similar composition in a steady magnetic field [5]. In order to justify the occurrence of this FOMP, K_3 should be negative ($K_3 < 0$). In figure 4 we present the magnetic phase diagram for a uniaxial ferromagnet with $K_1 < 0$ in the $(K_2/K_1, K_3/K_1)$ plane. The equations of the boundary lines and a detailed description can be found in [6]. The bold line in this figure represents the qualitative path of the relative values of the anisotropy constants of $Nd_3(FeTi)_{29}$ in this magnetic phase diagram as the temperature decreases. The first crossing of the boundary, planar



to conical

takes place at T_{SR}. As the temperature decreases, the FOMP starts to appear and consequently we are getting into the region between the 1–o′ lines in which the approach to saturation along a direction in the basal plane is through a FOMP

(that is an easy-cone magnetic structure with a relative minimum in the free energy within the basal plane). The reported prediction is consistent with all the experimental results. An additional comment concerning the shoulder found in χ_{AC} at $T = 150$ K is required; this could be explained in terms of the large increase of K_1 below this temperature (see figure 3).

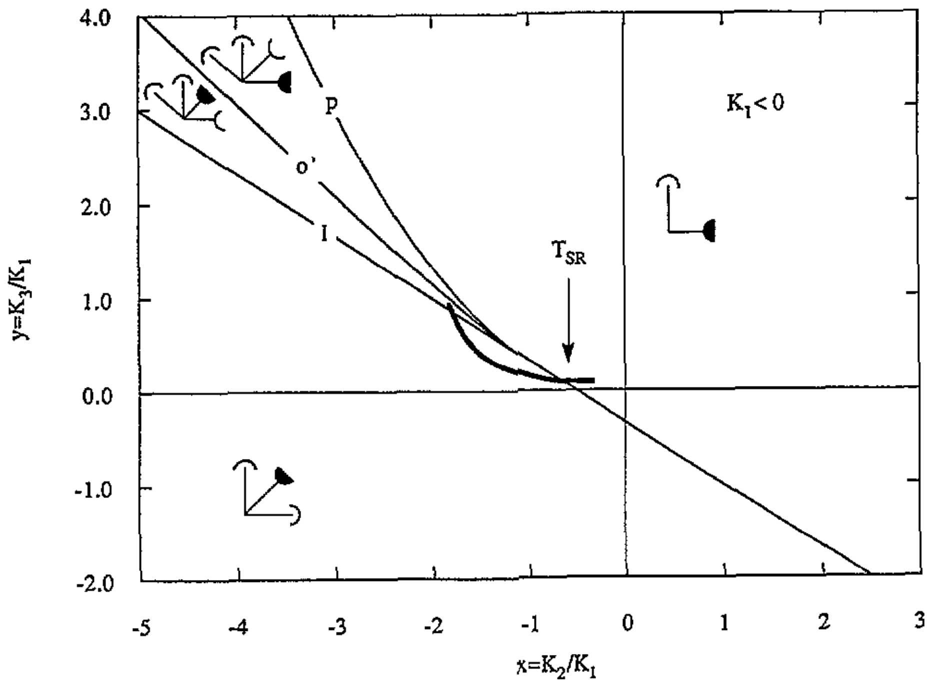


Figure 4. The magnetic phase diagram for a uniaxial ferromagnet [6] for $K_1 < 0$. The bold line represents the path of the K_1, K_2, K_3 relative values projected on the $(K_2/K_1, K_3/K_1)$ plane for decreasing temperatures.

In this letter we have presented thermal expansion, χ_{AC}, and SPD measurements on the new ternary compound $Nd_3(FeTi)_{29}$. From the interpretation of these experimental results we can reach the following conclusions.

(i) The large spontaneous magnetovolume effect observed at T_C ('invar'-like behaviour) is related to a strong enhancement of the Fe–Fe exchange interaction, being produced by a strong dependence of the exchange integral on distance. Additional experiments on χ_{AC} under pressure are under way, in order to confirm such a conclusion.

(ii) Two well defined magnetic regions have been found in this compound. Between $T_C = 413$ K and $T_{SR} = 233$ K the system is easy plane. At T_{SR} a spin reorientation transition takes place and the easy magnetization direction evolves towards an easy-cone magnetic structure.

(iii) The thermal dependence of the anisotropy fields H_A and H'_A measured by SPD can be explained following a path in the K_1, K_2, K_3 space in which $K_1 < 0$, $K_2 > 0$ and $K_3 < 0$ at any temperature, and the relative values of K_2/K_1 and K_3/K_1 evolve in such a way that the final state at low temperature (70 K) is such that the approach to saturation along the a-direction in the easy plane takes place through a $PC1$-type FOMP process.

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