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R. Verhoef, J. Franse, A. Menovsky, R. Radwa ski, Ji Song-Quan, et al.. HIGH-FIELD MAGNETI-SATION MEASUREMENTS ON R2Fe14B SINGLE CRYSTALS. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-565-C8-566. 10.1051/jphyscol:19888256 . jpa-00228424

HAL Id: jpa-00228424 https://hal.science/jpa-00228424

Submitted on 4 Feb 2008

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HIGH-FIELD MAGNETISATION MEASUREMENTS ON R₂Fe₁₄B SINGLE CRYSTALS

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Abstract. - High-field magnetisation measurements on single crystalline samples of $Pr_2Fe_{14}B$, $Nd_2Fe_{14}B$ and $Dy_2Fe_{14}B$ are presented and discussed within a two-sublattice model. Special attention is given to the high-field part of the curves $(B_0 > 20 \text{ T})$.

1. Introduction

Since the discovery in 1983 of the excellent magnetic properties of the $R_2Fe_{14}B$ family of compounds many research groups have made a large effort to elucidate its intriguing intrinsic magnetic properties. These properties are governed by the interplay between the crystalline-electric field (CEF) and the exchange interactions. In order to deduce the parameters describing these interactions a detailed study of the magnetisation curves of all the $R_2Fe_{14}B$ compounds is needed. For this study single crystals and high-magnetic fields are essential.

2. Samples and experimental techniques

The single crystals of $Pr_2Fe_{14}B$ (26 mg) and $Dy_2Fe_{14}B$ (19 mg) have been produced in the Laboratoire Louis Néel, Grenoble, France whereas the $Nd_2Fe_{14}B$ single crystal (sphere of 3 mm; 105 mg) has been produced in Amsterdam. In both cases a Czochralski technique was used.

High-field magnetisation measurements, at 4.2 K, were performed in the High-Field Installation of the University of Amsterdam [1]. Measurements were carried out by means of step-wise pulses with field intervals that can be programmed. For a more precise determination of the magnetisation curves, desirable in case of transitions, pulses in which the field increases or decreases linearly in time are employed.

3. Results and discussion

The magnetisation curves are analysed with the following phenomenological expression for the free energy:

 $E_{R-T} = E_a^R + E_a^T - \mu_0 n_{RT} M_T \cdot M_R - M_T \cdot B_0 - M_R \cdot B_0$ where M_T and M_R denote the magnetisation of the iron and rare-earth sublattice respectively, and where $\mathbf{B}_0 = \mu_0 \mathbf{H}$. The anisotropy energy E_a is expressed in terms of the Legendre functions, P_n^m , and the corresponding anisotropy coefficients, κ_n^m , which can be transformed into the CEF coefficients A_n^m . The 3d-4f exchange interaction is expressed in a molecularfield approximation where n_{RT} is the intersublattice molecular-field coefficient. For the iron anisotropy energy E_a^{T} and the iron magnetic moment M_{T} , the values derived for the Y₂Fe₁₄B compound are used. A more detailed description of this type of analysis is given elsewhere [2].

 $Nd_2Fe_{14}B.$ – In figure 1 the experimental magnetisation curves of $Nd_2Fe_{14}B$ are presented together with the calculated fits. From the experimental data we derive a value for the spontaneous magnetisation of 192 Am²/kg, a tilt angle of 30.8° and a transition in the [100] direction at an applied field of 16.3 T. These results are in good agreement with the data reported before [3, 4, 5].



Fig. 1. – High-field magnetisation curves of $Nd_2Fe_{14}B$ at 4.2 K. Dotted lines represent fits to the experimental data.

In case the field was applied along the [110] direction we went to the ultimate field limit of our installation and measured up to 40 T. No transition is found up to this field value along this direction and the magnetisation is still not saturated.

The fit to the experimental data results in the following CEF parameters (in Ka_0^{-n}): $A_2^0 = +330$, $A_4^0 =$ -16, $A_6^0 = -2.9$, $A_4^4 = +3.9$, $A_6^4 = -12$ and in a value of the molecular field experienced by the Nd moment ($= \mu_0 n_{\rm RT} M_{\rm T}$) of about 350 T, which is a measure for the exchange interaction. These values are in satisfying agreement with data reported by Cadogan *et al.* [5].

 $Pr_2Fe_{14}B$. – The magnetisation curves of $Pr_2Fe_{14}B$ are presented in figure 2. Our experimental results differ from the results reported by Hiroyoshi *et al.* [6]. Firstly, we derive a somewhat lower value for the spontaneous magnetisation of 185 Am²/kg and somewhat higher values for the transition fields of 13.2 T and 17.1 T along the [100] and [110] direction, respectively. Secondly, we observe an intersection of the [100] and [110] curves after the transitions which is not present in the curves of Hiroyoshi *et al.*



Fig. 2. – High-field magnetisation curves of $Pr_2Fe_{14}B$ at 4.2 K. Full lines and markers represent experimental data.

A satisfying fit with reasonable values for the CEF and exchange parameters has not yet been achieved. After the transitions the magnetisation does not reach the saturation value. This indicates that there is a minimum in the free energy for a direction which is tilted away from the c axis by about 55° . This minimum is mainly created by the CEF parameter B_6^0 . However, the observed reduced magnetisation at high fields, $B \ge 25$ T, hampers the fitting procedure. This reduction may be related to field induced non-collinear magnetic structures. When forcing the magnetisation by means of the applied magnetic field to be in the basal plane, the anisotropy energy connected with the off-diagonal CEF parameters can be so large that the Pr moments at the two different rare-earth sites (f and g) can be split up into four sublattices.

 $Dy_2Fe_{14}B$. – The magnetisation curves of the $Dy_2Fe_{14}B$ sample are presented in figure 3. The es-

sential point to note in these magnetisation curves is the intersection of the hard [100] and [110] axis curves with the easy axis [001] curve at 30 T. This intersection reflects the bending towards each other of the two sublattice moments under the influence of the increasing magnetic field. For the molecular field experienced by the Dy moment we derive, from our fit to the experimental data a value of 137 T. The crystal field parameters derived from the fit are (in Ka_0^{-n}): $A_2^0 =$ +285, $A_4^0 = -14$ and $A_6^0 = 0.5$, values which are somewhat different from data reported by Givord *et al.* [7]. In addition, the angle ε by which the moment configuration deviates from the strict antiparallel one, is determined as a function of the applied field. It is found that in a field of 10 T ε is already 6° increasing to 18° for 35 T.



Fig. 3. – High-field magnetisation curves of $Dy_2Fe_{14}B$ at 4.2 K. Full lines and markers represent experimental data.

Acknowledgements

This work is supported by the Commission of the European Communities within the CEAM programme.

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