



# Magnetic properties of $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$ : Suppression of exchange interactions and magnetocrystalline anisotropy by Co substitution

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## ARTICLE INFO

### Article history:

Received 26 November 2017

Received in revised form

8 January 2018

Accepted 9 January 2018

Available online 13 January 2018

### Keywords:

Ferrimagnetism

Exchange interactions

Magnetic anisotropy

Field-induced transition

## ABSTRACT

In  $3d-4f$  intermetallic compounds, a Co substitution for Fe usually strengthens the exchange interactions due to a shift of the Fermi energy to a region of the  $3d$  band with higher density of states. Here, we study the influence of Co on the magnetism of ferrimagnetic  $\text{DyFe}_5\text{Al}_7$  using magnetization measurements in static (up to 14 T) and pulsed (up to 58 T) magnetic fields. We find that the homogeneity range of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  is limited to  $x \leq 2.5$ . The Co substitution for Fe produces a strong detrimental effect on the  $3d-3d$  intrasublattice exchange interactions, which leads to a pronounced decrease of the Curie temperature and of the  $3d-4f$  intersublattice exchange field. A reduction of the density of states at the Fermi level might occur due to a broadening of the  $3d$  band. A decrease of the rare-earth contribution to the magnetic anisotropy energy is also observed with increasing Co content. This is attributed to a competition between the Fe and Co contributions to the crystal electric field at the Dy site.

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## 1. Introduction

Intermetallic compounds based on  $3d$  ( $T$ ) and  $4f$  ( $R$ ) elements are of interest for the following reasons. Firstly, some of them display good favorable magnetic properties and attract much attention due to their potential to be used as materials for permanent magnets. Secondly, the  $3d$  and  $4f$  electrons are involved in fairly complex interactions leading to physics that requires fundamental understanding. The competition of the intra- and intersublattice exchange interactions together with the crystal electric field gives rise to spontaneous and field-induced phase transitions. Analysis of these phenomena provides information on these fundamental magnetic properties and may lead to approaches to control them.

In the past, the most widely used methods to tune magnetic properties of  $R-T$  compounds have been application of pressure [1–3], interstitial introduction of H, C, and N [4–8], and elemental substitution [9–13]. It is often very challenging to predict how the

magnetic properties change as a function of tuning parameter, even qualitatively. However, when substituting Fe with Co, it may usually be assumed that the magnetic ordering temperature of a given material will increase, at least for relatively low Co concentrations. This behavior has been observed for various materials and is usually explained to be due to changes in the filling of the  $3d$  band (see, e.g., Refs. [13–19]).

Intermetallic compounds  $RT_{12-y}M_y$  crystallizing in the tetragonal  $\text{ThMn}_{12}$ -type crystal structure can be stabilized by the addition of a third nonmagnetic element,  $M$ . Typically, a rather small concentration of  $M$  is needed, e.g.,  $y \approx 1$  for  $RT_{12-y}\text{Ti}_y$  for both,  $T = \text{Fe}$  and  $\text{Co}$  [20–23]. Aluminum-stabilized compounds exist with  $T = \text{Fe}$ , but do not form with  $T = \text{Co}$ .  $R\text{Fe}_{12-y}\text{Al}_y$  requires a larger amount of Al and exist within the homogeneity range  $6 \leq y \leq 8$  [24–30]. The Fe sublattice shows competing exchange interactions. It is ordered antiferromagnetically for high Al concentration,  $7 \leq y \leq 8$ . By contrast, for  $y \approx 6$ , the Fe sublattice is ferromagnetic. The type of order changes in the vicinity of  $y \approx 7$  [31]. Ferromagnetic order within the Fe sublattice can be stabilized by interaction with the magnetic rare-earth atoms [32].

It is known that for  $RT_{12-y}M_y$  compounds, the rare-earth

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contribution to the magnetic anisotropy energy has opposite signs for  $T = \text{Fe}$  and  $T = \text{Co}$  due to the different crystal electric fields [33]. For the Fe-based compounds, it was also found that different Fe sites contribute with different types to the magnetic anisotropy [31]. This indicates that  $RT_{12-y}M_y$  displays also competing anisotropy interactions. It is reasonable to assume that an elemental substitution can modify the exchange interactions and magnetic anisotropy considerably.

A partial substitution of Fe with Co is possible for  $R\text{Fe}_5\text{Al}_7$ . A recent study of  $\text{HoFe}_{5-x}\text{Co}_x\text{Al}_7$  showed a detrimental effect of such a substitution on both, the exchange interactions and magnetic anisotropy [34]. This surprising finding motivates us to examine the magnetism of other members of the  $R\text{Fe}_5\text{Al}_7$  family where Fe is substituted by Co.

In the present work, we report on magnetization measurements of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  ( $x = 0-2$ ) compounds. Due to their large magnetic anisotropy, a detailed study can only be performed on single crystals. We prepared samples with  $x = 0, 0.5, 1, 2$ , and  $2.5$ . The sample with  $x = 2.5$  was found to be not single phase. Therefore, we measured the magnetization of the single crystals with  $x = 0, 0.5, 1$ , and  $2$  in static and pulsed magnetic fields.

## 2. Experimental details

$\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  single crystals with  $x = 0, 0.5, 1$ , and  $2$  were grown by a modified Czochralski method from high-purity elements (Dy 99.9%, Fe and Co 99.99%, Al 99.999%) in a tri-arc furnace using tungsten electrodes on a water-cooled copper crucible under protective Ar atmosphere. The pulling speed was 20 mm/h. The resulting ingots had a cylindrical shape with a height of approximately 30 mm and a diameter of 5 mm. Part of the samples was ground into powder and standard x-ray diffraction analysis was performed to check the crystal structure and determine lattice parameters. Back-scattered Laue-diffraction patterns were used to confirm the single-crystalline state of the samples and orient them for magnetization measurements.

The magnetization was measured between 2 and 300 K in static magnetic fields up to 14 T using a Physical Property Measurement System (PPMS-14). The magnetization in pulsed magnetic fields up to 58 T was measured at the Dresden High Magnetic Field Laboratory using a magnetometer similar to that described in Ref. [35]. Absolute values of the magnetization measured in pulsed fields were obtained using the static-field data.

## 3. Results

Our x-ray diffraction analysis showed that all  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  samples for  $x$  between 0 and 2 are single phase and crystallize in the tetragonal  $\text{ThMn}_{12}$ -type crystal structure. We also tried to grow a single crystal with  $x = 2.5$  and found that this sample contained another phase besides the  $\text{ThMn}_{12}$ -type phase. This suggests that the compounds for  $x > 2.5$  do not form, which is in accordance with a previous work on  $\text{HoFe}_{5-x}\text{Co}_x\text{Al}_7$  [34]. Therefore, we conclude that the homogeneity range of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  is limited to  $x \leq 2.5$ .

Fig. 1a shows the concentration dependence of the lattice parameters,  $a$  and  $c$ , of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$ . Parameter  $a$  decreases from 869 pm for  $x = 0$  to 863 pm for  $x = 2.5$ , whereas parameter  $c$  stays constant at about 504 pm. This suggests that the  $3d$  atoms form bonds rather in the basal plane, than along the  $[001]$  axis. Such an anisotropic shrinkage of the lattice results in a 1% decrease of the unit-cell volume,  $V$  (Fig. 1b). The  $c/a$  ratio grows linearly with increasing Co content. The decrease of the unit-cell volume is attributed to a smaller metallic radius of Co (1.25 Å) as compared to that of Fe (1.26 Å).

Fig. 2 shows field-dependent magnetization measured for fields

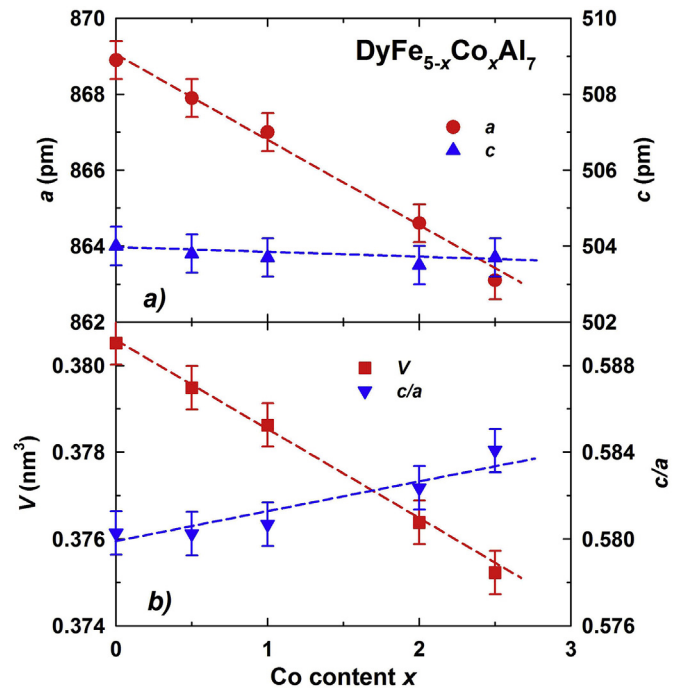


Fig. 1. Concentration dependence of (a) the lattice parameters,  $a$  and  $c$ , and (b) volume of the elementary cell,  $V$ , and the ratio  $c/a$  of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$ .

applied along the principal crystallographic directions of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  single crystals at 2 K (the data are only given for decreasing field, hysteresis will be discussed below). For all samples, there is a nonzero spontaneous magnetic moment along the  $[100]$  and  $[110]$  axes and no spontaneous component along the  $[001]$  axis. Therefore, the magnetic moments lie in the basal plane. Anisotropy is also observed within the basal plane, the  $[110]$  axis is the easy magnetization direction (Refs. [32,36,37] give incorrect data on the easy magnetization direction of  $\text{DyFe}_5\text{Al}_7$  since the  $[100]$  and  $[110]$  axes were interchanged). For the spontaneous magnetic moments along the  $[100]$  and  $[110]$  directions,  $M_s^{100}$  and  $M_s^{110}$ , respectively, the relation  $M_s^{100}/M_s^{110} \approx \cos 45^\circ$  holds, which reflects a good quality of the single crystals and their proper orientation. The anisotropy between the  $[100]$  and  $[110]$  directions is observed up to high temperatures, which can be seen, e.g., by the difference between the  $M(T)$  dependences for the compounds with  $x = 0, 0.5$ , and  $1$  (Fig. 3). The fact that the  $M(T)$  dependences for the  $[100]$  and  $[110]$  axes coincide just below the magnetic ordering temperature means that the anisotropy field is less than the applied field of 0.1 T.

As seen in Fig. 2, the spontaneous magnetic moments of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$  gradually increase with  $x$ . For example,  $M_s^{110} = 2.1 \mu_B/\text{f.u.}$  for  $x = 0$  and  $3 \mu_B/\text{f.u.}$  for  $x = 2$ . Fig. 4a shows that both,  $M_s^{100}$  and  $M_s^{110}$ , increase linearly with  $x$ . Taking into account that  $\text{DyFe}_5\text{Al}_7$  is a collinear ferrimagnet [27] with an almost fully developed Dy magnetic moment,  $M_{\text{Dy}}$ , the magnetic moment of the Fe sublattice is given by  $M_{\text{Fe}} = M_{\text{Dy}} - M_s = 10 - 2.1 = 7.9 \mu_B/\text{f.u.}$  This gives  $1.58 \mu_B/\text{f.u.}$  for the average magnetic moment per Fe atom. Since  $M_s^{110} = 2.3, 2.5$ , and  $3 \mu_B/\text{f.u.}$  for  $x = 0.5, 1$ , and  $2$ , respectively, we can estimate the average magnetic moment per Co atom to be  $1.1 \mu_B/\text{f.u.}$  for all compounds.

The Co substitution for Fe weakens the exchange interactions as can be seen from Fig. 3 that shows temperature dependences of the magnetization in a field of 0.1 T applied along the  $[100]$  and  $[110]$  axes of  $\text{DyFe}_{5-x}\text{Co}_x\text{Al}_7$ . At 2 K, a field of 14 T was applied and then reduced to 0.1 T so that the single crystals were in a single-domain

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