

Journal of Alloys and Compounds 317-318 (2001) 459-464

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

Magnetic properties of a $DyCo_{10}V_2$ single crystal

O. Tegus^{a,b}, Y. Janssen^a, E. Brück^a, A.A. Menovsky^a, F.R. de Boer^a, K.H.J. Buschow^{a,*}

^aVan der Waals-Zeeman Institute, University of Amsterdam, Amsterdam, The Netherlands ^bDepartment of Physics, Inner Mongolia Normal University, Huhhot 010022, Huhhot, PR China

Abstract

We have studied the magnetic properties of a single crystal of the $DyCo_{10}V_2$ compound. This compound has the tetragonal ThMn₁₂ structure. The easy magnetization direction is parallel to the *c* axis at high temperatures. The magnetization tilts away from the *c* axis below the spin reorientation temperature at T_{SR} =42 K, reaching a value of θ_C =27° at 5 K. The magnetic compensation of the Co and Dy sublattice magnetizations leads to a sharp minimum at T_{COMP} =118 K. The shape of the M(T) curves is strongly field-dependent and is considerably influenced by the thermal history of the sample. This behaviour is attributed to the development of a rather strong intrinsic coercivity at low temperatures, originating from the presence of narrow Bloch walls. The temperature dependence of the coercivity, diverging at the compensation temperature, was compared with model predictions. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: DyCo10V2; Magnetization process; Coercivity; Rare-earth-transition-metal compounds

1. Introduction

In a previous investigation made on polycrystalline material we showed that the easy magnetization direction of $DyCo_{10}V_2$ is parallel to the c axis at room temperature [1]. From measurements of the temperature dependence of the magnetization, it was derived that a spin-reorientation transition takes place at about 41 K and that there is a compensation temperature at about 120 K. We also measured magnetisation hysteresis loops on magneticallyaligned samples at various temperatures from 5 to 300 K with fields applied parallel and perpendicular to the alignment direction. For samples field-cooled to below the compensation temperatures, shifts of the loop centres into the range of negative magnetizations were observed. These features and the occurrence of negative remanences were explained in terms of the magnetization reversal when passing the compensation temperature upon cooling.

2. Experimental

The single crystalline rod of $DyCo_{10}V_2$ was grown by means of a modified tri-arc Czochralski technique. The sample cut from this rod was shown by X-ray diffraction to have the tetragonal ThMn₁₂ structure. EPMA examination showed that tiny parts on the periphery of the cut sample may have a deviating composition. For the present type of investigation we will assume that the influence of these impurities are of minor importance.

The magnetic measurements on the single crystal were made on a SQUID magnetometer in the temperature range 5-300 K in magnetic fields up to 5 T and in the Amsterdam High-field Installation in semicontinuous fields up to 35 T.

3. Results and discussion

The magnetic isotherms measured at 300 K, 200 K and 100 K in directions parallel and perpendicular to the cdirection are shown in Fig. 1a–c. All these measurements were made with increasing field strength. At these temperatures the coercivities are very small and do not interfere with the initial shape of these isotherms. It can be derived from the results shown in Fig. 1a–c that the easy direction is along the c direction at all three temperatures. The mutually antiparallel Dy and Co sublattice moments can bend towards each other in the applied field when the latter is perpendicular to the c direction. For this reason, the net moment may become higher in comparatively high fields than when measurements are made with the field applied parallel to the alignment direction. This may

^{*}Corresponding author.



Fig. 1. Magnetic isotherms measured at 300 K (a), 200 K (b) and 100 K (c) in directions parallel and perpendicular to the c direction. All these measurements were made with increasing field strength.

explain the crossing of the two isotherms shown in Fig. 1a and c.

As will be discussed in more detail below, coercivities of considerable magnitude are present at lower temperatures. For this reason we have measured the magnetic isotherms at 4.2 K in much higher field strengths and with decreasing field strengths in order to suppress any domain wall motion effects on the shapes of the isotherms. Results of these measurements are shown in Fig. 2. It is seen that in the low-field regime a moment component is present parallel as well as perpendicular to the c direction. This shows that the easy magnetization direction has tilted away from the c axis. When extrapolating the low-field parts of the isotherm to zero field, one may obtain the spontaneous moment components along and perpendicular to the c direction and use these values to calculate the cone angle $\theta_{\rm C}$ the easy direction makes with the c axis. The value of $\theta_{\rm C}$ estimated in this way equals 17°. This value is slightly lower than the $\theta_{\rm C}$ value derived from measurements of the angle dependence of the magnetization to be discussed below.

The hysteresis behaviour measured at 5 K is shown in Fig. 3. These results were obtained by cooling the orientated single crystal to 5 K in zero field and measuring first the virgin magnetization curve. The occurrence of a high coercivity in conjunction with the flat character of the virgin curve can be taken as a signature of the presence of narrow domain walls [2-5]. These narrow walls are strongly pinned on magnetic obstacles of atomic dimensions. The strong increase of the magnetization on the virgin curve at $\mu_0 H_{\rm P} = 2$ T marks the propagation field $H_{\rm P}$ at which the external field is able to detach the narrow walls from the pinning sites. For higher fields the walls are removed from the crystal. When decreasing the field from 5 T into the region of negative fields reversed domains and domain walls can be nucleated but their movement is impeded by the pinning sites so that the reversed domains



Fig. 2. Magnetic isotherms at 4.2 K measured in two directions with decreasing field strengths.



Fig. 3. Hysteresis loop measured at 5 K. The data were obtained by cooling the orientated single crystal to 5 K in zero field and measuring first the virgin magnetization curve.

cannot spread into the crystal. This becomes possible again only for negative fields equal in magnitude to the propagation field, meaning that the coercive field is equal in absolute value to the propagation field, $H_{\rm C} = H_{\rm P}$. Measurement made at several other temperatures reveal essentially the same behaviour. Results are displayed in Fig. 4 where it can be seen that the propagation fields are strongly dependent on temperature. The temperature dependence of $H_{\rm P}$ (or $H_{\rm C}$) of the single crystal is shown in Fig. 5. When starting at 2 K, the intrinsic coercivity first decreases with increasing temperature. It seems to diverge at around 120 K and at still higher temperatures continues to decrease. Before discussing this unusual temperature dependence of the coercivity we will first discuss the temperature dependence of the magnetization because both temperature dependences are strongly correlated.

Results of measurements of the temperature dependence of the magnetization made with fields of various strengths applied along the c direction of the crystal are shown in



Fig. 4. Field dependence of the magnetization measured after cooling the orientated single crystal to 5 K in zero field.



Fig. 5. Temperature dependence of the coercivity. The inset shows a plot of $H_{\rm C}^{-1}$ vs. *T*.

Fig. 6. Curve A was obtained after cooling the crystal to 5 K in zero field and measuring in a relatively small field of only 0.05 T. Under these conditions, the magnetization at 5 K can be represented as a point on the virgin curve. The sample is practically in the demagnetized state and the magnetization remains low up to about 180 K. At this temperature the coercivity has apparently dropped below the value of the measuring field and for temperatures higher than 180 K the sample can become magnetized by the measuring field. This is revealed by the strong increase of the magnetization with increasing temperature. This magnetized sample was subsequently cooled again to 5 K and measurements were repeated in higher field strengths. Under these conditions the magnetization at 5 K can be represented as a point on the upper branch of the hysteresis loop and the temperature dependence of the magnetization of curves B and C in Fig. 6 can be taken as free from any interference of domain walls. As done previously [1], we attribute the discontinuity at $T_{SR} = 42$ K to the occurrence



Fig. 6. Temperature dependence of the magnetization made with fields of various strengths applied along the c direction of the crystal. The thermal history of these samples is described in the main text.

of a spin reorientation and the sharp minimum at $T_{\text{COMP}} = 120$ K to mutual cancellation of the Dy and Co sublattice magnetization at this temperature.

Curve A in Fig. 7 was obtained with the same small measuring field (0.05 T) as curve A in Fig. 6, the only difference being that we magnetized the sample in 5 T before the measurements. At 5 K the magnetization corresponds to a point on the upper branch of the hysteresis loop and in the lower temperature range curve A in Fig. 7 shows the same features as curves B and C in Fig. 6. However, because the measuring field is still below the coercivity at T_{COMP} the total magnetization is not able to re-adjust itself according to the applied field when it has reversed its direction for $T > T_{\text{COMP}}$. The measured magnetization remains therefore negative until the measuring field becomes stronger than the coercivity at about 180 K. Curve B in Fig. 7 shows result obtained with a field of 1 T, similar to curve B in Fig. 6. The difference with the latter curve is that curve B in Fig. 7 was obtained by starting from the demagnetized state at 5 K. Inspection of the data in Fig. 3 shows that the magnetization in 1 T at this temperature still corresponds to a point on the virgin curve. This situation persists up to about 35 K. As can be seen in Figs. 4 and 5, the value of $H_{\rm C}$ has dropped to below the measuring field at this temperature. This means that increasing temperature leads to a strong jump in the magnetization value and above 35 K curve B in Fig. 7 can be regarded as representing points lying on the upper branch of the hysteresis loop. From thereon curve B in Fig. 7 is no longer different from curve B in Fig. 6. It follows from these results that a reliable shape for a curve representative for the temperature dependence of the spontaneous magnetization can only be derived from magnetization values corresponding to the upper branch of the hysteresis loop, like curves B and C in Fig. 6. These curves show that $M_{\rm s}(T)$ initially decreases with increasing temperature. After vanishing at T_{COMP} it increases again.



Fig. 7. Temperature dependence of the magnetization made with fields of various strengths applied along the c direction of the crystal. The thermal history of these samples is described in the main text.

We will now return to the temperature dependence of the coercivity shown in Fig. 5.

According to the model proposed by Barbara and Uehara [5], the temperature dependence of the coercivity can be described by the expression

$$1/H_{\rm C}(T) = 1/H_{\rm C}(0) + \alpha T$$
 (1)

where α is proportional to the spontaneous magnetization over the squared wall energy, $\alpha \propto M_s / \gamma^2$. The squared wall energy is proportional to the product of the average exchange energy and average anisotropy energy. The average exchange energy is composed of the Dy and Co intrasublattice contributions and the Dy-Co intersublattice contributions. In the lowest temperature region only the Dy intrasublattice exchange energy is expected to be temperature dependent and to lead to some decrease of the average exchange energy with increasing temperature. Also the average anisotropy energy is composed of Dy and Co sublattice contributions. In the lowest temperature range the latter can be regarded as temperature independent, whereas the Dy sublattice contribution is expected to fall off with increasing temperature. Qualitatively, one may expect therefore that γ^2 decreases with increasing temperature in the lowest temperature range. From the temperature dependences of curves B and C in Fig. 6 one may furthermore derive that also $M_{\rm s}$ decreases with increasing temperature in the lowest temperature range. Because in this range both $M_{\rm S}$ and γ^2 decrease with increasing temperature, the overall result may be that $\alpha \propto M_{\rm s}/\gamma^2$ is not much temperature dependent in the lowest temperature range. It means that one may expect $1/H_{\rm C}$ to vary linearly with temperature, a behaviour that is seen in Fig. 5 (inset) to be fairly well obeyed up to about 65 K.

It is relatively unimportant whether γ^2 becomes less or more temperature dependent above 65 K. Because of the presence of the compensation temperature where $M_s = 0$, one derives from Eq. (1) that H_c is expected to diverge at this temperature. This fact that is clearly revealed in the experimental data shown in Fig. 5.

Measurements of the dependence of the magnetization on the angle between the magnetization and the applied field are shown in Fig. 8. These measurements were made at room temperature. After fixing the crystal, the angle dependence of the magnetization was measured, starting the measurements along the c direction of the crystal. The observation of maxima at $\theta = 0^{\circ}$ and $\theta = 180^{\circ}$ and a sharp minimum at $\theta = 90^{\circ}$ is in concord with the results displayed in Fig. 1, showing that at room temperature the magnetic anisotropy is along the c direction. The measurements were repeated at 30.5 K after cooling the sample to this temperature in the presence of a magnetic field. In that case the data will be representative for the upper branch of the hysteresis loop. There will be no domain wall movements that could possibly interfere with the angle dependence of the magnetization. Results are shown in Fig. 8b. The shape of this curve can be interpreted as follows.



Fig. 8. Angle dependence of the magnetization at 300 K (a), 30.5 K (b) and 5 K (c).

Closer inspection of the data in Fig. 8b shows that there is a shallow maximum in the $M(\theta)$ curve at about $\theta = 16^{\circ}$. This fact can be taken as an indication that the easy magnetization direction corresponds to an easy cone with a cone half angle $\theta_{\rm C}$ of about 16°. This agrees with the observation that there is still a magnetization component present in the field direction at $\theta = 90^{\circ}$ and that this component vanishes at an angle $\theta = 90 + \theta_{\rm C} = 102^{\circ}$. The value derived from the latter angle is $\theta_{\rm C} = 12^{\circ}$, which is slightly smaller than the value $\theta_{\rm C} = 17^{\circ}$ derived from the location of the maximum. This difference will be discussed later.

A prominent difference between the $M(\theta)$ curves shown in Fig. 8a and 8b is that in the former $M(\theta)$ remains positive over the whole trajectory whereas in the latter $M(\theta)$ adopts negative values for $\theta \ge 102^\circ$. The reason for the difference in behaviour is the coercivity, which is high at low temperatures and low or absent at room temperature. When the field component along the magnetization direction of the crystal becomes negative, the high coercivity present at low temperatures prevents magnetization reversal, meaning that the measured magnetization adopts negative values. Results obtained at 5 K are shown in Fig. 8c. The $M(\theta)$ curve at this temperature is seen to be essentially the same as that obtained at 30.5 K. There is a shallow maximum at $\theta_{\rm C} = 29^{\circ}$ and the $M(\theta)$ curve adopts negative values at $90 + \theta_{\rm C} = 90 + 27 = 117^{\circ}$. Also for this temperature one finds that the $\theta_{\rm C}$ value derived from the $M(\theta)$ maximum is slightly larger than the value derived from the sign change of $M(\theta)$. The reason for this is probably, that the moments are statistically distributed over all angles of the cone $\theta_{\rm C}$ when starting the $M(\theta)$ measurements with the applied field parallel to the c direction. When the field starts to deviate from the c direction, only the moment direction on the cone that is parallel to the applied field becomes stabilized. From experimental results not presented here, we have obtained evidence that the magnetic anisotropy in the basal plane is small but certainly not negligible. For this reason, the initial torque experienced by the moments on the cone is very small for low θ values in applied fields of 0.05 T. This means that the redistribution of cone angles into the single direction parallel to the field proceeds fairly sluggishly, leading to a rather unsharp $M(\theta)$ maximum that is shifted slightly to higher θ values. If we take the angle where $M(\theta)$ passes through zero as more reliable for determining the cone angle, we find that it changes from $\theta_{\rm C} = 27^{\circ}$ at 5 K to $\theta_{\rm C} = 12^{\circ}$ at 30.5 K. Bearing in mind that the spin reorientation was found to start at 42 K, the temperature dependence of the cone angle behaves as shown in Fig. 9.

4. Concluding remarks

We have investigated the magnetic properties of a single crystal of the $DyCo_{10}V_2$ compound. The easy magnetization direction is parallel to the *c* axis at high temperatures. The magnetization tilts away from the *ac* axis below the spin reorientation temperature at $T_{SR} = 42$ K, reaching a value of $\theta_c = 27^\circ$ at 5 K. We found that the temperature



Fig. 9. Temperature dependence of the cone angle.

dependence of the magnetization shows several unusual features. The occurrence of an easy cone at low temperatures implies that the first-order anisotropy constant K_1 changes from positive to negative values at T_{SR} , an anisotropy change that is reflected by a step-like increase of M(T) when cooling to below T_{SR} . A further anomaly in the M(T) curves is due to the magnetic compensation of the Co and Dy sublattice magnetizations which leads to a

sharp minimum at $T_{\text{COMP}} = 118$ K. The shape of the M(T) curves are not only strongly field-dependent but depend also considerably on the thermal history of the sample. This behaviour is attributed to the development of a rather strong intrinsic coercivity at low temperatures originating from the presence of narrow Bloch walls. The present results, obtained on a fairly large single crystal, refute previous explanations of the high coercivity at 5 K in terms of small single domain particles. The most prominent result obtained in the course of the present investigation is the unusual temperature dependence of the coercivity. We showed for the first time that the presence of narrow Bloch walls leads to a divergency of the coercivity at the compensation temperature.

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