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Magnetic properties of $\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds

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The temperature dependence of the magnetization of $\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds with $x=0, 2, 4$, and 6 was measured in fields up to 5.0 T and in the temperature range from 4.2 to 300 K in a SQUID magnetometer. Compensation temperatures were observed for the compounds with $x=4$ and 6. The values of the R - T exchange interaction obtained by fitting the M - T curves on the basis of two-sublattice molecular-field theory are compared with the values obtained from high-field magnetization measurements on powder particles free to be oriented by the applied field. In all four compounds magnetic transitions are found in the ac-susceptibility measurements which were carried out between 4.2 K and room temperature. The types of magnetic anisotropy were investigated by magnetization measurements on samples that were magnetically aligned at room temperature.
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I. INTRODUCTION

$\text{RT}_{10}\text{Si}_2$ compounds (R =rare earth, T =Fe, Co, Ni) form in the tetragonal ThMn_{12} -type structure. Like in many other Fe-rich rare-earth intermetallic compounds, the Curie temperature can be strongly increased by partially substituting Co for Fe. This substitution can also be accompanied by changes of the magnetic anisotropy and magnetization of the T sublattices. In $\text{RT}_{10}\text{Si}_2$ systems, the Fe sublattice, according to the results on $\text{YFe}_{10}\text{Si}_2$, has an easy-axis magnetic anisotropy,^{1,2} and the Co sublattice, according to results on $\text{YCo}_{10}\text{Si}_2$, has an easy-plane magnetic anisotropy.^{3,4} The Curie temperature of $\text{YCo}_{10}\text{Si}_2$ is about 300 K higher than that of $\text{YFe}_{10}\text{Si}_2$.⁴ To study the influence of the Co sublattice on the magnetic properties of 1:12 compounds, we have investigated the magnetic properties of the series $\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds with $x=0, 2, 4$, and 6.

II. EXPERIMENTS

$\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds with $x=0, 2, 4$, and 6 were prepared in the same way as the $\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds reported on in Ref. 5. The temperature dependence of the magnetization of samples consisting of powder particles free to be oriented by the applied field was measured from 4.2 K to room temperature in external fields of 2 T.

In order to gather information on the type of magnetic anisotropy of the compounds at room temperature, x-ray-diffraction experiments were carried out both on magnetically aligned samples and on samples consisting of randomly oriented powder particles. Bulk samples were used in ac-susceptibility measurements which were done in the temperature range from 4.2 to 300 K. To study the easy-magnetization direction at different temperatures, the magnetization was measured in a SQUID magnetometer as a function of the angle between the alignment direction of the sample and the applied field of 1.0 T.

III. RESULTS

The temperature dependence of the magnetization of $\text{DyCo}_4\text{Ni}_6\text{Si}_2$ in a field of 2 T is shown in Fig. 1. A temperature-independent ferromagnetic impurity contribu-

tion of $0.53\mu_B/\text{f.u.}$ has been subtracted in accordance with our former results.⁵ From the results on $\text{DyCo}_4\text{Ni}_6\text{Si}_2$, where we were able to study the magnetization fully up to the Curie temperature, we can derive the molecular-field coefficients, n_{TT} , n_{DyT} , and n_{DyDy} by fitting the curve to the temperature dependence of the magnetization obtained in a two-sublattice molecular-field model. The following relations were used:

$$\mathbf{B}_{\text{Dy}}(T) = \mathbf{B} + n_{\text{DyDy}}\mathbf{M}_{\text{Dy}}(T) + n_{\text{DyT}}\mathbf{M}_{\text{T}}(T),$$

$$\mathbf{B}_{\text{T}}(T) = \mathbf{B} + n_{\text{TT}}\mathbf{M}_{\text{T}}(T) + n_{\text{DyT}}\mathbf{M}_{\text{Dy}}(T).$$

The temperature dependence of each sublattice moment is determined by the Brillouin function $B_J(x)$:

$$M_{\text{Dy}}(T) = M_{\text{Dy}}(0)B_{J_{\text{Dy}}}[\mu_{\text{Dy}}B_{\text{Dy}}(T)/k_B T],$$

$$M_{\text{T}}(T) = M_{\text{T}}(0)B_{J_{\text{T}}}[\mu_{\text{T}}B_{\text{T}}(T)/k_B T].$$

Because the magnetization of the Dy sublattice decreases faster with increasing temperature than does the T-sublattice magnetization, there will be a compensation temperature at which the difference between the two sublattice moments, and therefore also the critical field for breaking the antiparallel configuration, both approach zero.

FIG. 1. Temperature dependence of the magnetization of $\text{DyCo}_4\text{Ni}_6\text{Si}_2$ in 2 T. The circles represent the experimental results and the full curve corresponds to the calculated magnetization.

FIG. 2. (a) Calculated temperature dependence of the Dy- and T-sublattice moments in $\text{DyCo}_4\text{Ni}_6\text{Si}_2$. (b) Calculated temperature dependence of the angle between the Dy- and T-sublattice moments in $\text{DyCo}_4\text{Ni}_6\text{Si}_2$.

Therefore, in our calculation the temperature dependence of the angle between the two-sublattice moments has been taken into account. This angle, which determines the value of the magnetization, is found by minimizing the free energy:

$$F(T) = n_{\text{DyT}} M_{\text{Dy}}(T) M_{\text{T}}(T) \cos \alpha - B \sqrt{M_{\text{Dy}}^2(T) + M_{\text{T}}^2(T) + 2 M_{\text{Dy}}(T) M_{\text{T}}(T) \cos \alpha}.$$

The fit to the experimental magnetization curve is shown in Fig. 1 as a solid line. The molecular-field coefficients used in the fitting procedure are $n_{\text{DyDy}} = 1.1 \times 10^{23}$ Tf.u./J, $n_{\text{DyT}} = 4.9 \times 10^{23}$ Tf.u./J, and $n_{\text{TT}} = 7.8 \times 10^{24}$ Tf.u./J. The value for n_{DyT} has been taken equal to the value derived previously from high-field magnetization measurements.⁵ One can see in Fig. 1 that the magnetization above 130 K is not fitted very well. This may be due to the change of the magnetic anisotropy around this temperature which will be discussed below and/or to the crystallographic disorder of the T sublattice which may give rise to an inhomogeneous magnetization. In particular, the broadening of the magnetization around T_C may be attributed to this disorder.

The temperature dependences of the Dy- and T-sublattice moments and of the angle between M_{Dy} and M_{T} are shown in Fig. 2(b). Around the compensation temperature, the antiparallel configuration between M_{Dy} and M_{T} is broken, albeit only slightly [see insert of Fig. 2(b)]. At higher temperatures, the two sublattice moments remain antiparallel until the Curie temperature is reached.

FIG. 3. X-ray diffraction patterns at room temperature on a magnetically aligned $\text{YCo}_{10}\text{Si}_2$ sample (top) and a randomly oriented powder sample.

The magnetic anisotropy of the Co sublattice is of the easy-plane type. This is demonstrated in Fig. 3 which shows x-ray-diffraction patterns taken on a magnetically aligned $\text{YCo}_{10}\text{Si}_2$ sample with the alignment direction perpendicular to the surface and on a sample consisting of randomly oriented powder. One can see that for the aligned sample, the [002] reflection has disappeared whereas the [400] reflection is strongly enhanced.

Figure 4 shows the temperature dependence of the ac susceptibility measured on bulk samples between 4.2 and 300 K. All four compounds have clear magnetic transitions. The transition at about 270 K in $\text{DyCo}_4\text{Ni}_6\text{Si}_2$ is related to the Curie temperature which is consistent with the M - T measurements. The second transition in this compound is visible as a clear kink but is rather broad in the other compounds. The small anomaly in $\text{DyCo}_4\text{Ni}_6\text{Si}_2$ at low temperatures is probably due to a second phase.

To interpret the ac-susceptibility results, the magnetization of aligned samples was measured as a function of angle between the alignment direction and the direction of the ex-

FIG. 4. Temperature dependence of the ac susceptibility of $\text{DyCo}_{10-x}\text{Ni}_x\text{Si}_2$ compounds.

FIG. 5. Dependence of the magnetization of $\text{DyCo}_{10}\text{Si}_2$ on the angle between the alignment direction and the direction of the external field.

ternal field. The sample was mounted in such a way that the external field is along the direction in which the sample was aligned at room temperature. Therefore, at a certain temperature, the magnetization measured along the external field will get its maximum value when the easy-magnetization direction is in the field direction. The magnetization measured along the external field will get its minimum value when the hard-magnetization direction is in the field direction.

Figure 5 shows the experimental results for $\text{DyCo}_{10}\text{Si}_2$ measured at 220, 200, 190, 180, and 170 K. The curves have been normalized to the zero-angle magnetization values. The easy-magnetization direction of $\text{DyCo}_{10}\text{Si}_2$ can be seen to change at 190 K from easy plane above this temperature (established in the x-ray experiment) to easy axis below this temperature. At low temperatures, the magnetic anisotropy of the compound is dominated by the Dy-sublattice anisotropy. At high temperatures, the 3d-sublattice anisotropy (easy-plane type) becomes most important. Between about 150 and

250 K the two sublattice anisotropies are comparable. The competition between the two sublattices can clearly be seen in Fig. 5. At 190 K, the magnetization is nearly independent of the direction of the external field, indicating that the anisotropy has become very weak.

IV. DISCUSSION AND CONCLUSIONS

It is shown that Ni substitution for Co in $\text{DyCo}_{10}\text{Si}_2$ strongly influences the magnetic anisotropy. The ac susceptibility results show that the spin-reorientation temperature decreases with increasing Ni content (see Fig. 4). This may either be due to an increase of the T-sublattice anisotropy upon Ni substitution or to a decrease of the Dy-sublattice anisotropy upon Ni substitution or to both effects. This point will be the subject of further studies.

The molecular-field analysis of the magnetization shows that the Dy-Dy exchange interaction is almost zero in $\text{DyCo}_4\text{Ni}_6\text{Si}_2$ and that n_{DyT} is about one order of magnitude smaller than n_{TT} .

The analysis of the temperature dependence of the magnetization presented in this article shows that, in systems with a compensation temperature, when the external field is not very low, the angle between the two sublattice moments has to be taken into account.

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