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MAGNETIC ANISOTROPIC BEHAVIOUR OF RbNi$_{1-x}$Co$_x$F$_3$

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Résumé. — On a déterminé les valeurs à 87 °K de $K_1$, $K_2$ et de l'aimantation saturation des composés RbNi$_{1-x}$Co$_x$F$_3$ ($0 \leq x \leq 0.20$). L'aimantation saturation présente une anisotropie prononcée. Les variations de $K_1$ et $K_2$ en fonction de $H$ sont analogues à celles observées par Suits et al. A l'aide d'un modèle de champ cristallin à un ion on a calculé pour RbNiF$_3$ les constantes $K_1$ et $K_2$ et l'aimantation saturation. Pour celle-ci l'accord avec la valeur mesurée est bon, alors que pour $K_1$ et $K_2$ on obtient seulement l'ordre de grandeur. Pour RbNi$_{1-x}$Co$_x$F$_3$, la distribution des cations étant inconnue, on ne peut pas obtenir de résultats quantitatifs.

Abstract. — $K_1$, $K_2$ and the magnetic moment were measured on RbNi$_{1-x}$Co$_x$F$_3$ ($0 \leq x \leq 0.20$) at 87 °K by means of magnetization and torque curves. The magnetic moment shows a pronounced anisotropy. The $K_1$ and $K_2$ values as a function of $x$ show the same behaviour as observed by Suits et al. but the quantitative agreement is not good. By a one ion crystalline field model it is possible to calculate for RbNiF$_3$ $K_1$ and $K_2$ values which agree in sign and magnitude and $\mu$-values which agree quantitatively with the experimental values. For RbNi$_{1-x}$Co$_x$F$_3$ the unknown cation distribution does not allow quantitative calculations.

I. Introduction. — In the course of a study of the anisotropic magnetic properties of $d^5$-ions in magnetic compounds with various crystal structures we performed magnetization and anisotropy measurements on hexagonal fluorides of the composition

RbCo$_2$Ni$_{1-x}$F$_3$ ($0 \leq x \leq 0.2$)

in order to compare magnetic moment and anisotropy values with those obtained by a one ion crystalline field model. Single crystal samples were prepared either by the well-known combination of chemical reaction and flux melt [1]

$$3 \text{RbHF}_2 + (1-x)\text{NiCl}_2 \cdot x\text{CoCl}_2 =$$

$$= \text{RbNi}_{1-x}\text{Co}_x\text{F}_3 + 2 \text{RbCl} + 3 \text{HF}$$

or by the Bridgman-Stockbarger method. The Co : Ni ratio was determined by usual chemical analysis procedures. Magnetization and anisotropy measurements were made ballistically in different crystallographic directions and also by the torque method in the (1010) plane as a function of field strength at $T = 87$ °K. The maximum field strength for the magnetization measurements was 30 kOe and for the torque measurements 21 kOe.

II. Experimental Results. — The magnetization curves show besides a large crystal anisotropy a pronounced anisotropic behaviour of the magnetic moment especially for $x < 0.05$ and $x > 0.15$ (e.g. Fig. 1 for $x = 0$). Between $x = 0.05$ and $x = 0.15$ the anisotropic effects are smaller, due to the change of the easy magnetization axis from the a-plane to the c-axis. In order to interpret our measurements we start from the following expression for the free energy, assuming that the magnetic moment obeys the same symmetry relations as the magnetocrystalline energy [2, 3]:

$$\frac{1}{V} F = \frac{1}{V}(F_K + F_H) = K_0 + K_1 \sin^2 \theta + K_2 \sin^4 \theta - H M_{\text{sat}}(1 - k_1 \sin^2 \theta - k_2 \sin^4 \theta) \cos(\theta_0 - \theta) - \frac{1}{2} \chi H^2 \cos^2(\theta_0 - \theta)$$

(1)

$K_1$, $K_2$, $k_1$ and $k_2$ are the constants of magnetocrystalline anisotropy and anisotropy of magnetic moment resp., $\theta_0$ and $\theta$ the angle between c-axis and H or $M_x$ resp. and $\chi$ the differential susceptibility. By minimizing this expression with respect to $\theta$ we get the stable position of $M_x$ for a given $H$ and $\theta_0$ and therefrom we can calculate the magnetization and torque curves in the well-known manner.

Neglecting $k_2$ both the experimental magnetization curves and torque curves can be fitted very well by the calculated curves based upon the same values of $M_x$, $K_1$, $K_2$, $k_1$ and $\chi$. $M_x$ and $\chi$ change only by a small amount, from 74 to 76 G and from 0.17 to 0.20 $\times 10^{-3}$ resp., in the studied composition range.

Fig. 1. — Magnetization curves for RbNiF$_3$ at 87 °K.
The dependence of $K_1$, $K_2$ and $k_1$ from the Co-content is shown in figure 2. Our $K_1$- and $K_2$-values do not agree well with those obtained by Suits et al. [3]. The discrepancy in the anisotropy constants for $x < 0.05$ and $x > 0.15$ may be due to the corrections which are caused by taking into account an anisotropy of magnetic moment. In the intermediate region we get, contrary to Suits et al., a monotonous change of the easy direction from the a-plane to the c-axis. This transition is field dependent. From eq. (1) follows for the cone angle $\theta_c$

$$\sin \theta_c = \sqrt{\frac{-K_1 + HM_t k_1}{2(K_2 + HM_s k_2)}}.$$  

(2) 

Table I shows a comparison of calculated and measured $\theta_c$-values for $x = 0.12$ at 4 different field strengths.

<table>
<thead>
<tr>
<th>$H$ [kOe]</th>
<th>6.55</th>
<th>11.7</th>
<th>17.0</th>
<th>21.7</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\theta_c$, calc.</td>
<td>30.6°</td>
<td>25.3°</td>
<td>18.7°</td>
<td>10.4°</td>
</tr>
<tr>
<td>$\theta_c$, obs.</td>
<td>26.8°</td>
<td>22.8°</td>
<td>18.6°</td>
<td>12.8°</td>
</tr>
</tbody>
</table>

Table II

<table>
<thead>
<tr>
<th>$K_1$ [cm$^{-1}$/ion]</th>
<th>$K_2$ [cm$^{-1}$/ion]</th>
<th>$\mu_0$ [mB]</th>
<th>$\mu_\perp$ [mB]</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiI</td>
<td>1.160</td>
<td>-0.004</td>
<td>2.396</td>
</tr>
<tr>
<td>NiII</td>
<td>-0.905</td>
<td>0.026</td>
<td>2.366</td>
</tr>
<tr>
<td>$\text{RbNiF}_3$</td>
<td>$6.10 \times 10^5$</td>
<td>$4.46 \times 10^4$</td>
<td>21.6</td>
</tr>
</tbody>
</table>

$||$ and $\perp$ denotes parallel and normal to c-axis resp.

The values for RbNiF$_3$ were obtained by assuming that Ni = 1/3 NiI + 2/3 NiII and that the magnetic moments and therefore also $H_x, H_y$ are directed oppositely for the two kinds of Ni$^{2+}$ ions. Sign and magnitude for $K_1$ and $K_2$ and the absolute value as well as the anisotropy of magnetic moment are in very good agreement with the experimental values.

The same calculations were performed for Co$^{2+}$. The results obtained are in good qualitative agreement with experiment. For example the $K_1$-values of CoI and CoII have the opposite sign and are one order larger compared with Ni. Unfortunately, the exact distribution of Co$^{2+}$ ions on the two different lattice sites in RbNi$\text{Co}_x$CoF$_3$ is not yet known, so that it is not worth calculating values for $K_1$, $K_2$ and the magnetic moments in different crystallographic directions.

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References