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AXIAL ANISOTROPY IN THE TERBIUM-HOLMIUM SYSTEM

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Résumé. — On a étudié l’anisotropie magnétique axiale du système des alliages Ho-Tb, en mesurant la susceptibility magnétique de monocristaux, dans des champs jusqu’à 16 kOe, pour des températures comprises entre 80 et 700 K. A haute température l’inverse de la susceptibility varie linéairement avec la température mais une courbure se développe à mesure que l’on approche de la température de Néel. La constante d’anisotropie \( D \) de l’ion isolé est positive pour tout le système et varie légèrement d’une façon non linéaire avec la concentration. On a étudié la dépendance de la température sur cette variation. De plus, on a mesuré le nombre de magnétons de Bohr, la température de Néel et la température de Curie.

Abstract. — The axial magnetic anisotropy has been studied in the Ho-Tb alloy system, by measuring the magnetic susceptibility of single crystals in fields up to 16 kOe and at temperatures between 80 K and 700 K. At high temperatures the inverse susceptibility varies linearly with temperature, but a significant curvature develops as the Néel temperature is approached. The single ion anisotropy constant \( D \) is positive for the whole system and varies slightly non-linearly with concentration. The temperature dependence of this variation has been studied. Furthermore the variation of the effective Bohr magneton number, the Néel temperature and the Curie temperature have been measured.

1. Experimental results. — The magnetic properties of single crystals of Ho, Ho-10 \% Tb, Ho-50 \% Tb, Ho-90 \% Tb and Tb have been investigated between 80 K and 700 K in applied fields up to 16 kOe. Magnetic moment measurements were made along the a, b and c directions of the close-packed hexagonal structures with a Foner type vibrating sample magnetometer [1]. The magnetic properties of single crystals of Tb and Ho have been studied by the Ames group [2, 3] below room temperature and by Aléonard, Buto and Bloch [4] in the temperature range 0°C-100°C. Measurements below room temperature on single crystals of the alloys have been performed at Ames [5]. In the present work we report paramagnetic measurements from which the axial anisotropy constant can be extracted and its variation with alloy concentration studied.

The crystals were grown by a modified strain anneal method similar to that described by Nigh [6]. From these crystals 3 mm spheres were spark cut, oriented with x-rays and mounted on the magnetometer sample holder with an orientation better than 1°. The geometry of the apparatus was such that measurements along all directions in a crystal plane could be performed by simple rotation of the sampleholder. An anisotropy of less than 0.2 \% in this plane could be distinguished. Because of the high reactivity of these materials, the sample was mounted mechanically without glue in an argon atmosphere, for the high temperature measurements. The magnetic moment and the applied field could both be measured to within 0.5 \% and the temperature within 0.5 K.

The measurements in the ab-plane showed no anisotropy in the paramagnetic and antiferromagnetic regions. In the ferromagnetic region the b-axis was the easy axis. The measurements in the bc-plane showed anisotropy. This is illustrated in figure 1, where the inverse initial susceptibility in the c and b directions are plotted against temperature for a representative sample. The susceptibility for the b-direction is always greater than that for the c-direction. The inverse susceptibility varies linearly with temperature at high temperatures but a significant curvature develops as the Néel temperature is approached. This is most marked in the Ho rich alloys. The anisotropy is a decreasing function of temperature as illustrated in figure 2, where \( 1/T_{\text{c}} - 1/T_b \) is plotted against temperature for all the samples. For pure Ho this temperature dependence is in agreement with that due to thermal dilatation in a point charge model but for Tb and all the alloys the anisotropy temperature dependence is up to 6 times larger than that expected on this model. It seems likely that improvements on the single-ion molecular field model will be required to explain this discrepancy.

Figure 3a shows a plot of \( T_{N} \).

\[
T_C \text{ and } \theta_p = \frac{2\theta_b + \theta_c}{3}
\]
The difference in inverse initial susceptibility for the Ho-Tb alloy system against concentration. The antiferromagnetic region is widened and enhanced at the expense of the ferromagnetic region, as Ho is added to Tb.

The appropriate Hamiltonian for the $4f$ localized moment system is:

$$\mathcal{H} = - \sum_{m,n} J(R_m - R_n) S_m \cdot S_n + \sum_{n} \{ D_n J_{zn}^2 + E_n J_{zn}^4 + F_n J_{zn}^6 + G_n (J_{zn}^4 + J_{zn}^6) \}$$

where the first sum describes the Ruderman-Kittel exchange interaction between spins via polarization of the conduction electrons. The second sum is the anisotropic energy resulting from interaction of the single ion total angular momentum with the crystalline electric field. Using this Hamiltonian with $E = F = G = 0$ we derive, in the molecular field approximation, the paramagnetic susceptibility for a binary alloy with concentrations $c_1$ and $c_2$.

$$\chi = \frac{3k}{\mu_B^2} \left\{ \frac{3}{k} \left[ T - \frac{2J(0)}{3k} \left( c_1 x_1(g_1 - 1)^2 + c_2 x_2(g_2 - 1)^2 - c_1 c_2 x_1 x_2(g_1 - g_2)^2 \right) \right] \right\}$$

Index 1 and 2 refer to the two components, $k = $ Boltzmann's constant, $\mu_B = $ Bohr magneton, $g = $ Landé factor, $x = J(J+1)$ and $J(0) = \frac{1}{2} J \langle \bar{R} \rangle$. Interpreting the experimental inverse susceptibility in terms of these formulae, but rewriting the anisotropy terms with a single value of $D$ obtained by equating $D_1$ and $D_2$, $\mu_{\text{eff}}$, $J(0)$ and $D$ are derived. In figure 3b $\mu_{\text{eff}}$ is plotted against concentration. As Ho is added to Tb, $\mu_{\text{eff}}$ has the theoretical value $[c_1 g_1^2 x_1 + c_2 g_2^2 x_2]^4$ until about 50% Ho and then rises 1.5% above it in the Ho-rich alloys. $J(0)$ is constant and equal to $2.8 \pm 0.1$ meV/atom over the whole system. This lack of variation is to be expected from the similarity of the conduction electron configuration over the alloy system. $D$ is positive over the whole system and shows a slight curvature with concentration as illustrated in figure 3c.

If we simply assume the anisotropy constants $D_1$ and $D_2$ to be equal to the pure metal values respectively and calculate $D$ from an appropriate weighted average we find the variation of $D$ with concentration which is also shown in figure 3c.

It should be noted that $D$ measured in this way includes contributions from the higher order anisotropy constants in the form

$$D_{\text{eff}} = D + \frac{F}{7} (6 J^2 + 6 J - 5) +$$

$$+ \frac{E}{7} (5 J^4 + 10 J^3 - 5 J^2 - 10 J + 7)$$

for a pure metal. This can be seen by using the total anisotropy Hamiltonian.

References