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MAGNETIZATION AND MAGNETOSTRICTION OF TERBIUM-SCANDIUM ALLOYS

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Résumé. — Nous avons mesuré le paramètre cristallin, l’aimantation, la dilatation thermique et la magnétostriction d’une série d’échantillons monocristallins et polycristallins d’alliage Tb-Sc. Les diverses données expérimentales sont examinées en référence à la théorie de Callen et Callen et l’énergie magnéto-élastique est évaluée pour les diverses compositions.

Abstract. — Lattice parameters, magnetization, thermal expansion and magnetostriction of a series of single crystal and polycrystalline specimens of Tb-Sc alloys have been measured. The various experimental observations are examined in terms of the single ion theory of Callen & Callen and the magnetoelastic energy evaluated for the various compositions.

I. Introduction. — Scandium forms solid solutions with the heavy rare earths which have been studied by the Oak Ridge [1] group using neutron diffraction techniques. The Gd-Sc series has also been examined magnetically [2]. Scandium stabilises the helical antiferromagnetic state and depresses both Néel and Curie points. We have studied the Tb-Sc series in greater detail.

II. Experimental details. — Measurements of magnetization, thermal expansion and magnetostriction along the principal crystallographic directions have been made on single crystal discs of Tb$_{0.89}$Sc$_{1-x}$ where $x = 0.89, 0.825$ and $0.695$. These were produced by Metals Research Ltd. by annealing the alloy at $\sim 1300 \, ^\circ\text{C}$ and spark machining the ingot to give discs of diameter $\sim 5 \, \text{mm}$ and thickness $\sim 1 \, \text{mm}$. Offcuts were used for analysis. Measurements were also made on polycrystalline samples with $x = 0.5$ and $0.25$, prepared by repeated argon arc melting of weighed quantities of material.

Lattice parameters at room temperature were determined by the X-ray powder method with Cohen’s [3] corrections for systematic errors. Density was determined w. r. t. C$_6$H$_5$Br. Homogeneity was checked by X-ray fluorescence spectrometry. This method and conventional spectroscopy were used to check the purity. Absorption spectroscopy gave better estimates of composition in certain samples. Alloy compositions are accurate to within $\pm 0.5$ atomic % and homogeneity was good. The sample purity was of the order $4 \, \text{N}$, excluding dissolved gases.

A Foner type magnetometer, calibrated against pure Iron using the data of Weiss and Forrer [4], was used for magnetization measurements between $4.2 \, ^\circ\text{K}$ and $300 \, ^\circ\text{K}$. Thermal expansion and magnetostriction were measured using strain gauges in a D. C. bridge with a photoelectric galvanometer amplifier as null detector. The strain measurements were checked against the thermal expansion data of Nix and MacNair [5] for Copper. Strains of order $5 \times 10^{-7}$ were detectable.

III. Results and discussion. — The alloys have h. c. p. structure. As seen in figure 1, the a-axis parameters follow Vegard’s Law within experimental accuracy. The c-axis parameter, however, shows a negative deviation, while the pycnometric density exhibits a corresponding positive deviation. The pycnometric and X-ray densities agree within experimental error.

Two discs of Tb$_{0.89}$Sc$_{0.11}$ alloy were used, one with the basal plane in the plane of the disc and the other with the c-axis and one of the b-axes in the plane. Magnetization was measured as a function of applied fields (up to $9 \, \text{kOe}$) over the temperature range along a, b and c directions, the former being the easy direction and the c-axis very hard. Saturation

![Figure 1 - Variation of lattice parameter and density with composition.](http://dx.doi.org/10.1051/jphyscol:1971179)
magnetization at 4.2 °K was found by extrapolating the ς versus \( H^{-1} \) curve to \( H = \infty \). To estimate the ferromagnetic Curie point \( \theta_f \) the linear portion of the \( \sigma^2 \) versus \( T \) curve was extrapolated to \( \sigma^2 = 0 \). The value of \( T \) so obtained was plotted against \( H \) and extrapolation to \( H = 0 \) gave \( \theta_f \). The \( T \) versus \( H \) plot showed the variation with temperature of the critical field required to switch from helical antiferromagnetism to ferromagnetism. The Néel point is also field dependent. Above this the susceptibility follows a Curie-Weiss law. From the linear \( \chi \) versus \( T \) plots, the paramagnetic Curie temperature \( \theta_p \) and the effective moment \( \mu_{\text{eff}} \) have been estimated. The basal plane and the c-axis values of \( \theta_p \) differ indicating high anisotropy. The polycrystalline \( \theta_p \) is well represented by \( \frac{1}{2} \theta_p \) (c-axis) + \( \frac{1}{3} \theta_p \) (basal). The variation of critical field with temperature and some representative magnetization data are shown in figure 2.

The \( \text{Tb}_{0.825}\text{Sc}_{0.175} \) crystals showed similar behaviour to the previous alloy, but with different values of the parameters. The other alloys do not show ferromagnetism, but have a characteristic Néel point except for \( \text{Tb}_{0.25}\text{Sc}_{0.75} \), which shows no ordering. The various parameters measured are summarised in Table I.

Thermal expansion measurements on \( \text{Tb}_{0.89}\text{Sc}_{0.11} \) single crystals show that the a-axis contracts from 300 °K down to \( \theta_f \) with a change of gradient around \( T_N \). At \( \theta_f \) there is a sharp contraction and in the ferromagnetic state the contraction continues. The c-axis contracts down to \( T_N \) after which expansion starts under the influence of inter-layer magnetic forces.

Scandium alloys do not obey the \( G^{2/3} \) law \((G = \text{de Gennes Factor})\) like the Yttrium alloys. Using available data and our measurements it appears that \( T_N \) varies as \( G^{4/3} \) while \( \theta_N \) varies approximately as \( G^2 \). From the room temperature magnetization, the susceptibility per unit mass of Terbium in each alloy was estimated. This follows an expression of the form \( \chi^{-1} = \chi_0^{-1}(1 - \alpha N) \), where \( \alpha \) is a constant and \( N \) the Tb atomic fraction, as observed in Tb-Y alloys by Weinstein and al. [7]. Here, too, the susceptibility of Tb at infinite dilution in \( \text{Sc}(x_0) \) leads to a value of \( \mu_{\text{eff}} = (10.2 \pm 0.1) \mu_B \), significantly higher than that for Tb.

The saturation moments per Tb atom in the two Tb rich alloys were also higher than in pure Tb. It is suggested that the presence of Sc increases the conduction electron polarisation and hence the observed moments.

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**Table 1**

<table>
<thead>
<tr>
<th>Alloy</th>
<th>( \theta_p ) °K</th>
<th>( T_N ) °K</th>
<th>( \theta_f ) °K</th>
<th>( \mu_{\text{eff}}/\text{Tb ion} )</th>
<th>( \mu_{\text{eff}}/\text{Tb atom} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{Tb}<em>{0.89}\text{Sc}</em>{0.11} )</td>
<td>195 ± 1</td>
<td>190 ± 2</td>
<td>197 ± 1</td>
<td>10.2 ± 0.2</td>
<td>9.6 ± 0.2</td>
</tr>
<tr>
<td>( \text{Tb}<em>{0.825}\text{Sc}</em>{0.175} )</td>
<td>186 ± 1</td>
<td>170 ± 2</td>
<td>75 ± 2</td>
<td>9.9 ± 0.2</td>
<td>9.4 ± 0.2</td>
</tr>
<tr>
<td>( \text{Tb}<em>{0.60}\text{Sc}</em>{0.395} )</td>
<td>—</td>
<td>130 ± 2</td>
<td>152 ± 2</td>
<td>9.7 ± 0.2</td>
<td>—</td>
</tr>
<tr>
<td>( \text{Tb}<em>{0.25}\text{Sc}</em>{0.75} )</td>
<td>—</td>
<td>63 ± 2</td>
<td>95 ± 2</td>
<td>10.3 ± 0.2</td>
<td>—</td>
</tr>
<tr>
<td>( \text{Tb}<em>{0.25}\text{Sc}</em>{0.3} )</td>
<td>—</td>
<td>15 ± 2</td>
<td>—</td>
<td>10.1 ± 0.2</td>
<td>—</td>
</tr>
</tbody>
</table>
No data exists for the elastic constants of these alloys and those for pure rare earths are not complete. We have tried to estimate the elastic constants from those of the elements. A crude estimate has been made of the constant \( C'' = 2(C_{11} - C_{12}) \). Using the relation \( E_{ms} = -C''(12)^2/8 \) (Cooper [8]), the magnetostrictive energy at 0 °K is about 1.25 °K/atom for Tb\(_{0.89}\)Sc\(_{0.11}\) and 0.8 °K/atom for Tb\(_{0.825}\)Sc\(_{0.175}\) indicating a rapid fall from the value of 2 °K/atom estimated for pure Tb. This rapid fall seems to account for the absence of any driving force for the transition to ferromagnetism even at high Tb concentration. From the experimental data, estimates of interplanar exchange constants and their variation with temperature have also been made. Wollan [9] discussed the effect of the smaller atomic volume of Scandium on the magnetic ordering. Our results are in line with Wollan's analysis indicating that the magnetism is dominated by the reduction in size due to alloying with Scandium.

**References**