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Magnetic susceptibility of dilute alloys of rare earths in magnesium

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Résumé. — Nous étudions le champ cristallin dans les alliages dilués des terres rares lourdes Tb, Dy, Ho, Er ou Tm dans le Mg par des mesures de susceptibilité, et d'aimantation. On a déduit le champ cristallin sur les impuretés d'erbium des résultats obtenus jusqu'à présent. La comparaison avec les champs cristallins sur les impuretés d'Er dans le Sc, Y ou Lu, qui sont aussi des métaux hexagonaux, indique que les champs cristallins sont remarquablement insensibles à la matrice métallique.

Abstract. — Crystal fields in dilute alloys of the heavy rare earths Tb, Dy, Ho, Er or Tm in Mg are currently being studied by susceptibility and magnetization measurements. From the data obtained so far, the crystal field on Er solutes has been deduced. Comparison to the crystal fields on Er solutes in Sc, Y, and Lu, which are also hcp metals, shows that the crystal fields are remarkably insensitive to the host.

The crystal fields on trivalent rare earth ions in the h.c.p. metals Sc, Y and Lu have been reported earlier [1]. In order to obtain information on the origin of the crystal field, it would be valuable to study other h.c.p. metals with different electronic structures. Magnesium is very suited to this as it has a different valence (2 instead of 3).

Single crystals of Magnesium with the rare earths Tb, Dy, Ho, Er or Tm of concentrations between 0.05 and 0.5 at. % were prepared using the Bridgman method. Starting materials were Magnesium, which we sublimed to obtain a concentration of unwanted magnetic impurities below 5 ppm, and rare earths of purity 99.9 %.

The crystals were checked for homogeneity by X-ray fluorescence; the concentrations were determined chemically. From the crystals spheres of 3 mm diameter were spark-cut. These samples were oriented by X-ray diffraction before mounting in the magnetometers.

The initial susceptibilities of the various rare earth solutes in different crystallographic directions were determined in the temperature range 1.5-300 K. This was done by measuring the magnetization in a low field \(0.134 \times 10^6 \text{ A m}^{-1}\) using the Faraday method. To avoid remanent fields the magnetizing field and gradient field were produced by a copper coil system immersed in liquid nitrogen.

In the analysis of the experimental data higher lying \(J\)-multiplets can be neglected. Within the lowest multiplet the Hamiltonian for one rare earth ion consists of the crystal field and Zeeman terms

\[
\mathcal{H} = \mathcal{H}_\text{CF} + \mathcal{H}_Z
\]

\[
\mathcal{H}_\text{CF} = B_{20} O_{20}^2(J) + B_{40} O_{40}^0(J) + B_{60} O_{60}^0(J) + B_{80} O_{80}^0(J)
\]

\[
\mathcal{H}_Z = g_J \mu_B J \cdot (\mathbf{H} + \lambda \cdot \mathbf{M})
\]

where \(O_i^m\) are the Stevens operators, \(B_m\) are the crystal field parameters, \(\mathbf{H}\) is the internal magnetic field, \(\lambda\) is the molecular field tensor (whose only nonzero components are \(\lambda_{bb} = \lambda_{bb} = \lambda_{cc} = \lambda_{\parallel}\)) and \(M\) is the magnetic moment per rare earth ion.

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Inverse susceptibilities vs. temperature for three Mg-Er alloys. Note that the data for Mg-0.256 at. % Er and Mg-0.227 at. % Er have been lowered 0.5 and \(1.0 \times 10^6 \text{ A m}^{-1}/\mu_B\), respectively.
In the dilute alloys studied exchange interactions are small perturbations on the crystal field and the exchange can be adequately treated in the molecular field model. The effect of the exchange on the low-field inverse susceptibility is a simple shift according to the formula

\[ \chi_i^{-1} = \chi_{\text{CF},i}^{-1} - \lambda_{ii} \quad (i = a, b, c) \quad (2) \]

where \( \chi_{\text{CF}} \) is the pure crystal field susceptibility.

Previous experience with other alloy systems \([1]\) shows that in alloys with Tb, Dy or Ho ordering effects may be present at low temperatures, making additional measurements necessary. Here we concentrate on Er alloys. A full report on all the alloys will be published later.

The experimental initial susceptibilities of Er in three different alloys are illustrated in figure 1. From the slope in the region 60-200 K we determined the concentrations, which agree with the chemical analysis within the uncertainty of the latter. Least-squares fits were made to obtain the crystal field parameters \( B_{20}, B_{40}, \) and \( B_{60} \) while keeping \( B_{66} \) equal to \( 0.0078 \times B_{60} \).

The resulting parameter sets for the three alloys agree within the uncertainty of their determination. Only for the alloy of the highest concentration significant non-zero exchange parameters were detected.

\[ \lambda_1 = (-0.024 \pm 0.01) \times 10^6 \text{ A m}^{-1} \text{ at.}/\mu_B; \]
\[ \lambda_9 = (-0.05 \pm 0.01) \times 10^6 \text{ A m}^{-1} \text{ at.}/\mu_B. \]

For the two alloys of lower concentration exchange parameters were set equal to zero. Subsequent fitting of all four crystal field parameters gave a slightly larger value of \( B_{66} \) (\( \sim 10.9 \times B_{60} \)) with only minor changes in the other parameters. All the curves in figure 1 have been calculated using the average set of crystal field parameters given in table I.

The results in ref. \([1]\) for both heavy and light rare earth solutes have shown that \( |B_{20}| \) decreases strongly with the \( c/a \)-ratio, whereas the higher order parameters are approximately independent of the lattice parameters. This can be inferred as Sc, Y, and Lu have similar electronic structures. As shown in table I the crystal fields in Mg fit into this scheme, indicating that the crystal fields are insensitive to the electronic structure of the metal.

<table>
<thead>
<tr>
<th>Host</th>
<th>Mg</th>
<th>Sc</th>
<th>Lu</th>
<th>Y</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c/a)_{ideal}</td>
<td>-c/a</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.008</td>
<td>0.041</td>
<td>0.050</td>
<td>0.062</td>
</tr>
<tr>
<td>( B_{20} ) (K)</td>
<td>-5 ± 3</td>
<td>-29 ± 3</td>
<td>-55 ± 7</td>
<td>-111 ± 13</td>
</tr>
<tr>
<td>( B_{40} ) (K)</td>
<td>5.5 ± 2</td>
<td>8.2 ± 2</td>
<td>9.6 ± 2</td>
<td>13.5 ± 6</td>
</tr>
<tr>
<td>( B_{60} ) (K)</td>
<td>13.5 ± 1</td>
<td>18.1 ± 2</td>
<td>16.1 ± 2</td>
<td>12.0 ± 3</td>
</tr>
<tr>
<td>( B_{66}/B_{60} )</td>
<td>10.9</td>
<td>10.2</td>
<td>10.4</td>
<td>11.1</td>
</tr>
</tbody>
</table>

References
