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CRYSTAL FIELD ANALYSIS OF THE MAGNETOCRYSTALLINE ANISOTROPY IN THE $R_2Fe_{14}B$ SERIES OF COMPOUNDS

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Abstract. – Analysis of the single crystal magnetization curves of $Pr_2Fe_{14}B$ measured from 4.2-275 K in fields of up to 18 T, and of the spin reorientation in $Yb_2Fe_{14}B$ reveal that there is a reduction of about 50% in the second-order rare earth crystal field compared to the rest of the series (Nd-Tm). The reduction is attributed to incipient valence instability in these two compounds.

The crystal field and exchange model [1] developed to account for the magnetic properties of the $R_2Fe_{14}B$ family of compounds and successfully applied to compounds with $R = \text{Nd, Tb, Dy, Ho, Er and Tm}$ [2] is now used to analyse the magnetic properties of $Pr_2Fe_{14}B$ and $Yb_2Fe_{14}B$.

We have measured the single-crystal magnetization curves of $Pr_2Fe_{14}B$ as a function of temperature for the three principal directions in fields of up to 18 T (Fig. 1) at the SNCI, Grenoble. The results were fitted using the rare earth – iron exchange coefficient $n_{PrFe} = 372 \mu_0$, deduced from the temperature dependence of the spontaneous magnetization, (compared to $307 \mu_0$ found for $Nd_2Fe_{14}B$). The crystal field coefficients used in the fit are listed in table I. The fourth-order terms are very small but of opposite sign to those for the rest of the series [1, 2]. Note in particular that the calculations reproduce the type-II first order magnetization processes (FOMP) below 100 K in both hard directions. These FOMPs have been measured independently by Hiroyoshi et al. [3] but an adequate fit was not obtained.

Table I. – Crystal field coefficients for $Pr_2Fe_{14}B$.

<table>
<thead>
<tr>
<th>$A_{20}$</th>
<th>$A_{42}^5$</th>
<th>$A_{40}$</th>
<th>$A_{40}^5$</th>
<th>$A_{40}^7$</th>
<th>$A_{40}^9$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K_0^{-2}$</td>
<td>$K_0^{-4}$</td>
<td>$K_0^{-6}$</td>
<td>$K_0^{-8}$</td>
<td>$K_0^{-10}$</td>
<td>$K_0^{-12}$</td>
</tr>
<tr>
<td>4f</td>
<td>176</td>
<td>-116</td>
<td>3</td>
<td>-9</td>
<td>-7</td>
</tr>
<tr>
<td>4g</td>
<td>179</td>
<td>351</td>
<td>3</td>
<td>8</td>
<td>-7</td>
</tr>
</tbody>
</table>

The analysis for $Yb_2Fe_{14}B$ involved scaling the crystal field coefficients to reproduce the axis-plane reorientation temperature, $T_r = 115$ K [4] using the value of the rare earth-iron exchange field coefficient $n_{YbFe} = 133 \mu_0$, deduced by extrapolating values for the other heavy rare earth compounds [2]. A substantial reduction of the $A_{4s}$s obtained for $Nd_2Fe_{14}B$ [1] was required to reproduce the observed reorientation temperature.

In figure 2, values of the second order crystal field coefficient averaged over the two sites, $A_{20}$, are plotted for all the compounds that we have analysed. Values

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for Pr and Yb are distinctly anomalous while \(A_{20}\) for the others is constant to within \(\pm 8\%\). It is not reasonable to expect \(50\%\) anomalies in the crystal field coefficients for a series of isostructural compounds when the rare earths are in well defined \(R^{3+}\) states. So the data in figure 2 suggest a modification of the 4f configuration for Pr and Yb.

In metallic rare earth systems there is always a finite hybridization of the 4f electrons with the d band which is characterized by an energy \(V_5\). When \(|V_5|/\Delta E \approx 1\), where \(\Delta E\) is the energy necessary to promote an electron from the 4f\(n\) configuration to the Fermi level, one is dealing with a valence fluctuation system [5]. Valence anomalies tend to appear at the beginning of the 4f series where the binding energy of the 4f electrons is relatively low and the trivalent 4f\(n\) configurations are close in energy, \(\Delta E\) is small and some valence instability occurs. The phenomenon has been studied in great detail in Ce compounds [6], but there is now growing evidence that Pr also may have a mixed configuration in certain metallic systems [7-10].

An indication that \(R_2Fe_14B\) may not have a simple 4f\(^2\) configuration is provided by the a lattice parameter, the value for \(R_2Fe_14B\) is 0.01 \(\text{Å}\) less than expected by interpolation from La to Gd [11]. By comparison, a for \(Ce_4Fe_14B\) is reduced by 0.06 \(\text{Å}\).

Valence anomalies also occur for Eu and Yb where the 4f binding energy is large and the divalent 4f\(^{n+1}\) configuration lies below the Fermi level, due to the Hund's rule stabilization energy of the half full or full 4f shell [12]. In these cases the 4f\(^{n+1}\) and 4f\(^n\) 5d configurations are close in energy. Eu does not form the \(R_2Fe_14B\) phase and the Yb in \(Yb_2Fe_14B\) is essentially trivalent, as evidenced by the spin reorientation transition. Nevertheless, the low Curie temperature [4] and crystal field parameters suggest that here too the ion is close to valence instability.

The \(R_2Fe_14B\) series provides a unique opportunity for observing the systematic variation of crystal field parameters for different rare earths in the same structure. In order to confirm our analysis, measurements of photoemission spectra, polarized and inelastic neutron scattering or electronic specific heat should be carried out to look for evidence of valence instability at the beginning and end of the rare-earth series.

**Acknowledgment**

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