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Magnetic properties of R(Fe,Mn)\(_{11}\)Ti compounds

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High-field magnetization measurements have been performed on both free-powder samples and magnetically aligned samples of R(Fe,Mn)\(_{11}\)Ti (R=Y, Nd, Sm, Gd, Er) compounds at 4.2 K in a step field up to 28 T. The temperature dependence of ac susceptibilities of R(Fe,Mn)\(_{11}\)Ti has been measured in a temperature range from 4.2 to 800 K. It has been found that both the magnetic moment of the transition-metal sublattice and the Curie temperature of R(Fe\(_{1-x}\)Mn\(_x\))\(_{11}\)Ti compounds decrease with increasing \(x\). Spin reorientations occur in Er(Fe\(_{1-x}\)Mn\(_x\))\(_{11}\)Ti compounds as the result of competition between the uniaxial anisotropy of the transition-metal sublattice and the planar or conical anisotropy of the Er sublattice. The inter-sublattice exchange constants \(n_{RT}\) of Er(Fe\(_{1-x}\)Mn\(_x\))\(_{11}\)Ti compounds have been deduced by the high-field free-powder method. The effect of the substitution of Fe by Mn on the magnetic anisotropies of R(Fe,Mn)\(_{11}\)Ti with R=Y, Nd, Sm has been investigated. First-order magnetization processes take place in Sm(Fe\(_{1-x}\)Mn\(_x\))\(_{11}\)Ti compounds when the external field is applied perpendicular to the alignment direction. © 1996 American Institute of Physics. [S0021-8979(96)34808-3]

I. INTRODUCTION

In the early 1980s, many efforts have been made to produce iron-rich compounds with the ThMn\(_{12}\) structure by substituting Mn by Fe in RMn\(_{12}\) compounds. However, since the solid solubility of Fe in YMn\(_{12}\), Fe\(_x\) limits to \(x=8\), this substitution cannot lead to an Fe-rich compound. Recently, the Fe-rich R(Fe,Mn)\(_{12}\) compounds with the ThMn\(_{12}\) structure have been obtained by adding a small amount of a third element, such as Ti, V, Cr, Mo, W, and Al. So it is possible to study the effect of Mn substitution on the magnetic properties of Fe-rich rare-earth transition-metal compounds with the ThMn\(_{12}\)-type structure, in which the Mn is added as a fourth element to substitute part of the iron. In the present work, we have chosen RFe\(_{11}\)Ti as the starting compound, and prepared the RFe\(_{11-x}\)Mn\(_x\)Ti compounds with \(x=0, 1, 2, 3, 4\). The magnetic properties of these compounds have been investigated by high-field magnetization measurements up to 21 T at 4.2 K and ac-susceptibility measurements from 4.2 to 800 K.

II. SAMPLE PREPARATION

Polycrystalline RFe\(_{11-x}\)Mn\(_x\)Ti ingots with R=Y, Nd, Sm, Gd, and Er and \(x=0, 1, 2, 3, 4\) have been prepared by induction-melting appropriate amounts of pure materials of a purity of at least 99.99 wt %. The ingots were remelted two times to achieve homogeneity. Weight losses during the melting due to evaporation of the rare-earth element and Mn were compensated for by starting with an excess of 3 wt % R (with respect to the R content) and 5 wt % Mn (with respect to the Mn content). The as-cast ingots were wrapped in Ta foil and sealed in a pre-evacuated and then argon-gas-filled quartz tube, followed by annealing at 1273 K for three weeks and then water-quenched to avoid possible metallographic phase transitions during the cooling process.

The crystal structures of the samples were checked by x-ray diffraction. It was found most of the samples have the ThMn\(_{12}\)-type tetragonal structure and a small amount of \(\alpha\)-Fe exists as a second phase.

III. RESULTS AND DISCUSSION

The high-field measurements at 4.2 K were performed on free-powder samples of RFe\(_{11-x}\)Mn\(_x\)Ti compounds to study saturation magnetization and exchange interaction. For the high-field free-powder (HFFP) measurements, fine-powder (<40 μm) samples were loosely loaded into the sample holders. These particles can be considered as monocRYSTALLINE and are free to rotate in the external field so that the total magnetic moment of the particles is always parallel to the external field direction. The values for the saturation magnetization obtained in this way are tabulated in Table I. One can see that, for RFe\(_{11-x}\)Mn\(_x\)Ti (R=Y, Nd, Sm) compounds, the saturation magnetization in these compounds decreases upon Mn substitution. In the YMn\(_{12}\)-type structure, since Y is nonmagnetic, the magnetic moment of the transition-metal sublattice is equal to the saturation magnetization. In NdFe\(_{11-x}\)Mn\(_x\)Ti and SmFe\(_{11-x}\)Mn\(_x\)Ti, if we assume that the 4f electrons of rare-earth element are well localized, the moment of the transition-metal sublattice \((M_T)\) can be obtained by subtracting the free-ion moment of the rare-earth element \((M_R)\) from the saturation magnetization \((M_s)\). The obtained \(M_T\) values are listed in Table II. In GdFe\(_{11-x}\)Mn\(_x\)Ti and ErFe\(_{11-x}\)Mn\(_x\)Ti compounds, \(M_s\) first
TABLE I. Saturation magnetization of RFe_{11−x}Mn_{x}Ti compounds.

<table>
<thead>
<tr>
<th>x</th>
<th>Y</th>
<th>Nd</th>
<th>Sm</th>
<th>Gd</th>
<th>Er</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>20.0</td>
<td>21.7</td>
<td>19.6</td>
<td>12.2</td>
<td>10.8</td>
</tr>
<tr>
<td>1</td>
<td>15.4</td>
<td>17.2</td>
<td>16.4</td>
<td>9.3</td>
<td>6.6</td>
</tr>
<tr>
<td>2</td>
<td>12.5</td>
<td>15.1</td>
<td>10.2</td>
<td>4.0</td>
<td>2.4</td>
</tr>
<tr>
<td>3</td>
<td>7.4</td>
<td>13.7</td>
<td>7.5</td>
<td>2.6</td>
<td>1.8</td>
</tr>
<tr>
<td>4</td>
<td>4.2</td>
<td>6.9</td>
<td>4.7</td>
<td>2.7</td>
<td>4.6</td>
</tr>
</tbody>
</table>

TABLE II. Magnetic moments of the transition-metal sublattice in RFe_{11−x}Mn_{x}Ti compounds.

<table>
<thead>
<tr>
<th>x</th>
<th>Y</th>
<th>Nd</th>
<th>Sm</th>
<th>Gd</th>
<th>Er</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>20.0</td>
<td>18.4</td>
<td>18.9</td>
<td>19.2</td>
<td>19.8</td>
</tr>
<tr>
<td>1</td>
<td>15.4</td>
<td>13.9</td>
<td>15.7</td>
<td>16.3</td>
<td>15.6</td>
</tr>
<tr>
<td>2</td>
<td>12.5</td>
<td>11.8</td>
<td>9.4</td>
<td>11.0</td>
<td>11.4</td>
</tr>
<tr>
<td>3</td>
<td>7.4</td>
<td>10.4</td>
<td>6.8</td>
<td>9.6</td>
<td>7.2</td>
</tr>
<tr>
<td>4</td>
<td>4.2</td>
<td>3.6</td>
<td>4.0</td>
<td>4.3</td>
<td>4.4</td>
</tr>
</tbody>
</table>

HFFP measurements are usually used to get information about the intersublattice interaction in heavy-rare-earth transition-metal compounds. If the external field is so high that the collinear configuration of the magnetic moments of the rare-earth and transition-metal sublattices is broken, the coupling constant $n_{RT}$ can be obtained from the slope of the bending process if the anisotropy of at least one of the sublattices is negligible. In external fields up to 28 T, the bending process can be observed in ErFe_{11−x}Mn_{x}Ti compounds with $x = 1$, 2, and 3 (see Fig. 1). Hurley has measured the free-powder magnetization process of ErFe_{11}Ti and its first critical field is 35 T. In the present work, the first critical fields have been determined to be 23, 10.5, and 0 T for $x = 1$, 2, and 3, respectively. The coupling constants deduced from the slope in the bending section are 4.5, 4.0, and 4.5 T f.u./μB for $x = 1$, 2, and 3, respectively. It seems that Mn substitution has no significant influence on $n_{RT}$. However, no bending process is found in GdFe_{11−x}Mn_{x}Ti compounds. This may be due to the relatively large $n_{RT}$ in Gd compounds.

For all investigated RFe_{11−x}Mn_{x}Ti compounds, $M_T$ decreases monotonically with increasing $x$. If the mean Fe moment were not affected by substitution and the Mn ion were nonmagnetic, $M_T$ would decrease linearly by $M_T(x) = M_T(0)(11-x)/11$, where $M_T(x)$ and $M_T(0)$ are the saturation magnetizations of RFe_{11−x}Mn_{x} and RFe_{11}Ti, respectively. The experimental results obviously deviate from this line, which is similar to the case of R(Co,Mn)_{11}Ti compounds. One possible interpretation for the relatively fast decrease of $M_T$ could be that the magnetic moment of iron is affected by the surrounding of Mn atoms which might be coupled antiferromagnetically with the Fe moment. Another possible reason for this is that the Mn atom which is larger than Fe prefers to substitute the Fe atom on the 8i site where Fe has the largest moment in RFe_{11}Ti.
lower than room temperature, so we cannot prepare well-aligned samples of these compounds at room temperature. For NdFe$_{11}$Ti, the magnetization along the alignment direction is smaller than that perpendicular to the alignment direction. A possible interpretation for this may be that the weak easy-axis anisotropy of NdFe$_{11}$Ti at room temperature changes to an easy-cone anisotropy with a cone angle larger than 45° at 4.2 K. With increasing $x$, the magnetization parallel to the orientation direction becomes relatively large indicating that Mn substitution decreases the cone angle at low temperature or causes an easy-cone anisotropy at room temperature. Of more interest are the SmFe$_{11}$Ti compounds in which first-order magnetization process (FOMP)-like anomalies appear in the magnetization curves measured along the direction perpendicular to the orientation direction (Fig. 2). These anomalies are similar to the one found by Hu et al.$^7$ in SmFe$_{11}$Ti. The critical field, corresponding to the maximum of the first derivative of the magnetization curve, decreases upon Mn substitution. The critical fields in SmFe$_{11}$Ti and SmFe$_{10}$MnTi are 10.2 and 8.9 T, respectively. In SmFe$_{12}$Mn$_2$Ti, the anomaly becomes hard to distinguish because of the poor alignment of the powder at room temperature, and the critical field decreases to about 8 T. These anomalies may be due to field-induced first-order transitions or quasi-FOMPs.$^8$

The ac susceptibilities of RFe$_{11-x}$Mn$_x$Ti compounds were measured in the temperature range from 4.2 to 800 K. The obtained $T_c$ values are listed in Table III. $T_c$ decreases monotonically with increasing $x$. This is similar to the concentration dependence of $M_T$. Spin reorientations from easy axis at high temperature to easy cone or easy plane at low temperature were found in ErFe$_{11-x}$Mn$_x$Ti compounds. At $T_{SR}$ and above, the uniaxial iron anisotropy usually determines the magnetization direction but at lower temperatures the rare-earth anisotropy may be dominant. The $T_{SR}$ corresponds to the minimum of $d\chi'/dT$. The $T_{SR}$ for compounds with $x=0, 1, 2, 3, 4$ are 48, 50.5, 99.4, and 171.4 K, respectively. The decrease of $T_{SR}$ with increasing $x$ may imply that the uniaxial anisotropy of the transition-metal sublattice decreases upon Mn substitution.

In conclusion, both the magnetic moments of the transition-metal sublattice and the Curie temperatures of the RFe$_{11-x}$Mn$_x$Ti $(x=0, 1, 2, 3, 4)$ compounds decrease with increasing $x$. The introduction of Mn atoms, which are non-magnetic or possess a magnetic moment antiparallel to the Fe moments, gives rise to a decrease of the mean Fe moment. In RFe$_{11-x}$Mn$_x$Ti, the transition-metal sublattice possesses uniaxial anisotropy which decreases upon Mn substitution. In RFe$_{11-x}$Mn$_x$Ti $(R=Nd, Er)$, there exists competition between the uniaxial anisotropy of the transition-metal sublattice and the planar or conical anisotropy of the rare-earth sublattice, which gives rise to spin reorientations in the ErFe$_{11-x}$Mn$_x$Ti compounds. FOMP-like anomalies found in SmFe$_{11-x}$Mn$_x$Ti compounds indicate that the high-order anisotropies play an important role in the magnetization processes of these compounds.

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