Magnetic properties of R$_2$Co$_{15}$Al$_2$ compounds with R=Y, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm

Bao-gen Shen$^a$ Zhao-hua Cheng, Shao-ying Zhang, Jing-yun Wang, Bing Liang, Hong-wei Zhang, and Wen-shan Zhan

State Key Laboratory of Magnetism, Institute of Physics & Center for Condensed Matter Physics, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, China

(Received 16 September 1998; accepted for publication 2 December 1998)

An investigation of the structure and the magnetic anisotropy of R$_2$Co$_{15}$Al$_2$ (R=Y, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm) compounds has been performed by means of x-ray diffraction and magnetization measurements. X-ray diffraction shows the prepared compounds to be single phase, having the hexagonal Th$_2$Ni$_{17}$-type structure for R=Y, Ho, Er, and Tm and the rhombohedral Th$_2$Zn$_{17}$-type structure for R=Ce, Pr, Nd, Gd, Tb, and Dy. Except for the Ce compound, the unit-cell volumes of R$_2$Co$_{15}$Al$_2$ compounds decrease in accordance with the lanthanide contraction. Substitution of Al for Co in R$_2$Co$_{17}$ leads to a decrease of the saturation magnetization at 1.5 K and Curie temperature. The exchange-coupling constants $J_{Co-Co}$ and $J_{R-Co}$ have been calculated by using the method based on magnetic ordering temperature. It is found that the $J_{R-Co}$ has a small dependence on the R elements and is almost not affected by the Al substitution. The Ce compound is found to exhibit an anomalous lattice parameter and magnetic characteristic, which are relative to the mixed-valence behavior of the Ce ion. X-ray diffraction measurements on magnetically aligned R$_2$Co$_{15}$Al$_2$ powders show that the compounds with R=Pr, Nd, Gd, Tb, Dy, and Ho have an easy-plane type of magnetic anisotropy, whereas the compounds with R=Y, Ce, Sm, Er, and Tm exhibit an easy-axis type of magnetic anisotropy at room temperature. The compounds R$_2$Co$_{15}$Al$_2$ with R=Pr and Ho exhibit a spin-reorientation transition and the spin-reorientation temperature is found to be 531 and 431 K, respectively. A strong uniaxial anisotropy is observed in Sm$_2$Co$_{15}$Al$_2$ compound with a magnetocrystalline anisotropy field of 84 kOe at room temperature. © 1999 American Institute of Physics. [S0021-8979(99)05605-4]

I. INTRODUCTION

Since discovering Sm$_2$Co$_{17}$ magnets, binary R$_2$Co$_{17}$ (R=rare earth) and their substituted compounds have been extensively studied on their structural and intrinsic magnetic properties, especially the magnetocrystalline anisotropy (see, e.g., a review article by Strnat$^1$ and the papers cited therein as well as Refs. 2–7). A strongly uniaxial magnetocrystalline anisotropy is required to achieve high coercivity. Although they have high saturation magnetization and Curie temperature, it is hard to find practical applications for the R$_2$Co$_{17}$ compounds by themselves due to the unfavorable anisotropy. These compounds are either weak uniaxial (e.g., Sm$_2$Co$_{17}$) or planar (e.g., Y$_2$Co$_{17}$, Pr$_2$Co$_{17}$) anisotropy. However, many investigations have demonstrated that the overall anisotropy can be increased or a planar anisotropy can be changed to a uniaxial one by replacing Co with an appropriate proportion of other elements such as Ti, V, Cr, Mn, Fe, Ga, Al, Si, etc. in the R$_2$Co$_{17}$ compounds,$^1$–$^{15}$ as is the case in the R$_2$Fe$_{17}$ compounds in which partial substitution of Ga, Al for Fe leads to a change of the magnetocrystalline anisotropy from planar to axial.$^{16,17}$ The cobalt sublattice anisotropy in the R$_2$Co$_{17}$ compounds is proved to be easy-plane anisotropy.$^{18}$

In recent studies it was found that the substitution of non-magnetic atoms Ga and Al has a significant effect on the magnetocrystalline anisotropy of the Co sublattice, such as a study on Gd$_2$Co$_{17-x}$Al$_x$ and Gd$_2$Co$_{17-x}$Ga$_x$ compounds has demonstrated that the easy-magnetization direction of the Co sublattice changes from the basal plane to the c axis with increasing Al or Ga concentration at room temperature.$^{11,12}$ The spin-reorientation transitions at higher temperature were also observed in Al- and Ti-substituted R$_2$Co$_{17}$ compounds.$^{13,14}$ In previous investigations, the various elemental substitutions (such as Ga, Al, and Si) for Fe in the R$_2$Fe$_{17}$ compounds were systematically investigated.$^{19–29}$ The results indicate that the substitution of these non-magnetic atoms for Fe has a profound influence on the magnetic properties, especially on determining the easy magnetization direction of these compounds. As an extension of our previous work, we study the effect of Ga, Al, Si, etc. substitution on the structure and magnetic properties of the Co-based 2:17-type compounds. In this article, we report our study results on the influence of Al substitution for Co in R$_2$Co$_{17}$ compounds on the crystal structure, Curie temperature, saturation magnetization, magnetic-coupling strength between the $4f$ and $3d$ moments, and magnetocrystalline anisotropy.
II. EXPERIMENT

$R_2Co_{15}Al_2$ compounds with $R=$ Y, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm were prepared by arc melting the constituent metals in an argon atmosphere of high purity. The starting materials R, Co, and Al were at least 99.9% pure. The ingots were remelted three times to ensure homogeneity. The samples were then annealed in argon atmosphere at 1273 K for 2 days. Phase identification was made by powder x-ray diffraction and thermomagnetic measurements at low magnetic fields. X-ray diffraction measurements on powder samples were performed using Cu $K_\alpha$ radiation to determine the lattice parameters.

The oriented samples for the magnetocrystalline anisotropy studies were made by mixing well-ground fine powder with epoxy resin and then aligning the particles in a magnetic field of 10 kOe. The easy-magnetization direction was identified from the x-ray diffraction patterns of the magnetically aligned samples. Magnetization curves of the free powder and the oriented samples were measured by using an extracting-sample magnetometer in fields up to 65 kOe and by a superconducting quantum interference device (SQUID) magnetometer in fields up to 70 kOe. The Curie temperature for all compounds and the spin-orientation temperatures for the Pr and Ho compounds were determined from temperature dependence of magnetization measured in a magnetic balance or a vibrating-sample magnetometer with a field of 1 kOe. The magnetocrystalline anisotropy field $H_A$ was examined by the singular point detection (SPD) technique at room temperature.

III. RESULTS AND DISCUSSION

All compounds studied here were found to be single phase having a 2:17-type structure. The $R_2Co_{15}Al_2$ compounds with $R=$ Y, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm were found to possess the rhombohedral Th$_2$Zn$_{17}$-type structure and the compounds with $R=$ Y, Ho, Er, and Tm the hexagonal Th$_2$Ni$_{17}$-type structure. The room-temperature lattice constants $a$ and $c$, and the unit-cell volume $v$ of the compounds are given in Table I. Except for the Ce compounds, the unit-cell volumes of the compounds decrease in accordance with the lanthanide contraction, as shown in Fig. 1. In order to compare the volume of the hexagonal cells with the rhombohedral cells we have multiplied the former by 3/2.

An x-ray powder diffraction pattern with $2\theta$ between 20° and 80° at room temperature for Pr$_2Co_{15}Al_2$ compound is collected with Cu $K_\alpha$ radiation and the step-by-step scanning of 0.02°/3 s in order to study the site occupation of the Al atoms. The powder-diffraction pattern of Pr$_2Co_{15}Al_2$ is shown in Fig. 2. The measured intensities were analyzed using the Rietveld method implemented in the program RIETAN. We found that the lowest $R_{wp}$ (weighted pattern $R$ factor) value is 13.85%. One can find that the fitting values are satisfactory, consistent with the experimental data. We

![Figure 1](image1.png)

**FIG. 1.** Dependence of the room-temperature unit-cell volume $v$ of $R_2Co_{15}Al_2$ compounds on the $R$ atomic number. In order to compare the volume of the hexagonal cells with the rhombohedral cells we have multiplied the former by 3/2.

![Figure 2](image2.png)

**FIG. 2.** Output from Rietveld analysis of the x-ray diffraction pattern of Pr$_2Co_{15}Al_2$. The observed data are indicated by a solid circle and the calculated profile by the continuous line overlying them. The lower curve is the difference between the observed and calculated intensity at each step, plotted on the same scale at the bottom. The bars denote the calculated Bragg-reflected positions.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$a$ (Å)</th>
<th>$c$ (Å)</th>
<th>$v$ (Å$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.396</td>
<td>8.185</td>
<td>499.6</td>
</tr>
<tr>
<td>Ce$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.434</td>
<td>12.288</td>
<td>757.0</td>
</tr>
<tr>
<td>Pr$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.491</td>
<td>12.375</td>
<td>772.6</td>
</tr>
<tr>
<td>Nd$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.484</td>
<td>12.322</td>
<td>768.1</td>
</tr>
<tr>
<td>Sm$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.459</td>
<td>12.285</td>
<td>762.2</td>
</tr>
<tr>
<td>Gd$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.444</td>
<td>12.280</td>
<td>758.2</td>
</tr>
<tr>
<td>Tb$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.423</td>
<td>12.282</td>
<td>754.7</td>
</tr>
<tr>
<td>Dy$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.404</td>
<td>12.242</td>
<td>748.8</td>
</tr>
<tr>
<td>Ho$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.392</td>
<td>8.164</td>
<td>497.9</td>
</tr>
<tr>
<td>Er$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.368</td>
<td>8.193</td>
<td>496.8</td>
</tr>
<tr>
<td>Tm$<em>2$Co$</em>{15}$Al$_2$</td>
<td>8.369</td>
<td>8.170</td>
<td>495.6</td>
</tr>
</tbody>
</table>
have found that in Pr$_2$Co$_{15}$Al$_2$ the Al atoms completely avoid the 9$d$ site and prefer the 6$c$ and 18$h$ sites, which is in agreement with the neutron diffraction result observed in Nd$_2$Co$_{17-x}$Al$_x$. For Pr$_2$Co$_{15}$Al$_2$, about 20.2% of the Fe atoms on the 6$c$ site and 17.6% of the Fe atoms on the 18$h$ site are substituted by Al atoms, respectively.

The field dependence of magnetization of R$_2$Co$_{15}$Al$_2$ compounds measured at 1.5 K is shown in Fig. 3. The saturation magnetization was derived from $M$ vs $1/H^2$ plots by extrapolating $1/H^2$ to zero. The values of saturation magnetization are listed in Table II in two ways: as the moment per mass unit $M_s$ and as the moment per formula unit $m_s$. The dependence of the magnetic moment $m_s$ on the R atomic number is presented in Fig. 4. In the compounds containing light-rare-earth ions, the R moments and the Co moments are ferromagnetically coupled and the saturation moment $m_s$ can be expressed by $m_s = 15\mu_{Co} + 2\mu_{R}$, where $\mu_{Co}$ and $\mu_{R}$ are the average Co moment and the R moment, respectively. However, the compounds with heavy rare earth are ferrimagnetic and $\mu_s$ is expressed by $\mu_s = 15\mu_{Co} - 2\mu_{R}$. If we assume the R moments to be equal to their $R^{3+}$ free-ion values respectively, we can derive values for the Co moment $\mu_{Co}$ in the various compounds from the saturation moment $\mu_s$.

These values are also listed in Table II, where it can be seen that the $\mu_{Co}$ values in the Al-substituted compounds are lower than the Co free-atom moment (1.7 $\mu_B$). This indicates that substitution of Al leads to a decrease of the Co atom moment.

The Curie temperature $T_C$ of the R$_2$Co$_{15}$Al$_2$ compounds is determined from the temperature dependence of the magnetization measured in a field of 1 kOe. Examples of these measurements are shown in Fig. 5 for R$_2$Co$_{15}$Al$_2$ compounds with R=Ce and Tb. The values of $T_C$ obtained from these thermomagnetic curves for the R$_2$Co$_{15}$Al$_2$ compounds are also listed in Table II and the dependence on the R atomic number is presented in Fig. 6.

The Curie temperature of the rare-earth cobalt compounds is determined by the Co–Co, the R–Co, and the R–R interactions. It is possible to determine the intersublattice-coupling strength from the difference in magnetic-ordering temperature of compounds in which R has a magnetic moment and nonmagnetic, respectively. In the high-

![FIG. 3. Magnetization curves of R$_2$Co$_{15}$Al$_2$ compounds at 1.5 K.](image)

![FIG. 4. Dependence of the magnetic moment $\mu_s$ at 1.5 K of R$_2$Co$_{15}$Al$_2$ compounds on the R atomic number.](image)

<table>
<thead>
<tr>
<th>Compound</th>
<th>$T_C$ (K)</th>
<th>$M_s$ (Am$^2$/kg)</th>
<th>$\mu_s$ ($\mu_B$)</th>
<th>$\mu_{Co}$ ($\mu_B$)</th>
<th>$-J_{Co}/k$ ($R_2$Co$_{15}$Al$_2$)</th>
<th>$-J_{Co}/k$ ($R_2$Co$_{17}$)</th>
<th>EMD</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Y)$<em>2$Co$</em>{15}$Al$_2$</td>
<td>891</td>
<td>103.9</td>
<td>20.8</td>
<td>1.39</td>
<td>···</td>
<td>···</td>
<td>c axis</td>
</tr>
<tr>
<td>Ce$<em>2$Co$</em>{15}$Al$_2$</td>
<td>751</td>
<td>79.6</td>
<td>17.4</td>
<td>1.16</td>
<td>···</td>
<td>···</td>
<td>c axis</td>
</tr>
<tr>
<td>Pr$<em>2$Co$</em>{15}$Al$_2$</td>
<td>893</td>
<td>111.7</td>
<td>24.4</td>
<td>1.20</td>
<td>10.6</td>
<td>8.3</td>
<td>plane</td>
</tr>
<tr>
<td>Nd$<em>2$Co$</em>{15}$Al$_2$</td>
<td>888</td>
<td>107.4</td>
<td>23.6</td>
<td>1.14</td>
<td>8.6</td>
<td>7.2</td>
<td>plane</td>
</tr>
<tr>
<td>Sm$<em>2$Co$</em>{15}$Al$_2$</td>
<td>904</td>
<td>61.3</td>
<td>13.6</td>
<td>0.81</td>
<td>11.6</td>
<td>···</td>
<td>c axis</td>
</tr>
<tr>
<td>Gd$<em>2$Co$</em>{15}$Al$_2$</td>
<td>922</td>
<td>24.5</td>
<td>5.5</td>
<td>1.30</td>
<td>9.6</td>
<td>9.6</td>
<td>plane</td>
</tr>
<tr>
<td>Tb$<em>2$Co$</em>{15}$Al$_2$</td>
<td>913</td>
<td>13.3</td>
<td>3.0</td>
<td>1.40</td>
<td>9.9</td>
<td>8.0</td>
<td>plane</td>
</tr>
<tr>
<td>Dy$<em>2$Co$</em>{15}$Al$_2$</td>
<td>901</td>
<td>4.8</td>
<td>1.1</td>
<td>1.41</td>
<td>8.0</td>
<td>7.3</td>
<td>plane</td>
</tr>
<tr>
<td>Ho$<em>2$Co$</em>{15}$Al$_2$</td>
<td>898</td>
<td>4.0</td>
<td>0.9</td>
<td>1.39</td>
<td>8.4</td>
<td>7.0</td>
<td>plane</td>
</tr>
<tr>
<td>Er$<em>2$Co$</em>{15}$Al$_2$</td>
<td>903</td>
<td>20.5</td>
<td>4.7</td>
<td>1.51</td>
<td>9.4</td>
<td>6.9</td>
<td>c axis</td>
</tr>
<tr>
<td>Tm$<em>2$Co$</em>{15}$Al$_2$</td>
<td>887</td>
<td>26.2</td>
<td>6.0</td>
<td>1.33</td>
<td>12.4</td>
<td>6.5</td>
<td>c axis</td>
</tr>
</tbody>
</table>

*Taking from Ref. 34.
temperature limit of the mean-field approach, one may neglect the R–R interaction relative to the R–T and T–T interaction. From the corresponding values of the 3d spin \( S_T \) and the Curie temperatures of the isostructural compounds with nonmagnetic R elements such as Y, we can obtain the exchange-coupling constant \( J_{TT} \) and \( J_{RT} \) by using the standard mean-field formula:

\[
J_{TT}/k^2 = 3T_{C,R}/2Z_{TT}S_T(S_T+1),
\]

\[
J_{RT}/k^2 = 9T_{C,R}(T_{C,R} - T_{C,0})/4Z_{TR}Z_{TT}S_T(S_T+1) \times (g_R - 1)^2J_R(J_R + 1),
\]

where \( T_{C,R} \) and \( T_{C,0} \) denote the Curie temperatures of the compounds with a magnetic R element and nonmagnetic R element, respectively, \( k \) is the Boltzmann constant, \( g_R \) is the Lande factor, \( Z_{RT} \) is the number of nearest T atom neighbors of an R atom and the value of \( Z_{TR} \) is related to \( Z_{RT} \).

Here the Curie temperature of the \( Y_2Co_{15}Al_2 \) compound is used as \( T_{C,0} \), thus the value of the Co–Co exchange-coupling constant is obtained to be \( J_{Co-Co}/k = 143 \) K according to Eq. (1). Assuming that the value of \( J_{Co-Co} \) is constant in \( R_2Co_{15}Al_2 \), we can apply the \( J_{Co-Co} \) value of \( Y_2Co_{15}Al_2 \) to \( R_2Co_{15}Al_2 \) when determining the R–Co exchange-coupling constant \( J_{R-Co} \). The values of \( J_{R-Co} \) are presented in Table II, together with those of \( R_2Co_{15}Al_2 \) compounds for comparison. It can be seen from Table II that \( J_{R-Co} \) has a small dependence on R elements and is almost not affected by the Al substitution. Similar effects were observed in other 2:17-type cobalt-based compounds and also in 2:17-type iron-based compounds substituted by Ga or Al atoms. On the basis of band-structure calculations performed by Brooks et al., Jacobs et al. offered a better explanation which can be applied to explain the constant behavior of \( J_{R-Co} \) in various series of pseudobinaries or pseudoternaries. Since Brooks et al. showed that the overall 4f–3d coupling strength depends on the ratio of the average 3d and 5d spins there may be cancellation of the effect of both spin reductions, leaving the strength of \( J_{R-Co} \) almost unchanged. The Curie temperature of \( R_2Co_{15}Al_2 \) compounds is found to decrease upon Al substitution. This situation is dramatically different from that observed in \( R_2Fe_{17-x}M_x \) (M=Ga and Al) compounds where the Curie temperature first strongly increases with increasing Al or Ga concentration and goes through a maximum value, then decreases with \( x \). The reason for this difference is probably associated with the fact that the magnetic coupling between Co moments in metal systems is always ferromagnetic, independent of Co moment and structure, leaving no room for antiferromagnetic types of 3d moment interaction that could reduce the Curie temperature. Since the Co–Co exchange coupling constants are about one order of magnitude larger than the R–Co exchange coupling constants (\( J_{Co-Co}/k = 143 \) K, \( -J_{R-Co}/k = 10 \) ± 2 K for \( R_2Co_{15}Al_2 \)), the Curie temperature in Co-based intermetallics is mainly determined by the Co–Co exchange interactions. The Al substitution for Co in \( R_2Co_{17} \) increases Co–Co interatomic distance and greatly weakens the Co–Co exchange coupling, in consequence, leading to the decrease of the Curie temperature. It was observed that the magnetic moment as well as Curie temperature for \( Y_2Co_{15}Al_2 \) compounds is anomalously low in comparison with the values obtained from a simple extrapolation by the other \( R_2Co_{15}Al_2 \) compounds, as shown in Figs. 4 and 6, respectively. It can also be seen from Fig. 1 that the unit-cell volumes of the Ce compounds show an evident anomaly. These anomalies may be evidence of the mixed-valence behavior of the Ce ion in Ce compounds, as is the case in other Ce–T compounds.

The x-ray diffraction studies on the aligned powder samples of \( R_2Co_{15}Al_2 \) have shown that the \( R_2Co_{15}Al_2 \) compounds with \( R=Pr, Nd, Gd, Tb, Dy, \) and Ho have an easy-plane type of magnetic anisotropy, and the compounds with \( R=Y, Ce, Sm, Er, \) and Tm have an easy c-axis type of magnetic anisotropy at room temperature. For the compound \( Y_2Co_{17} \), the easy magnetization direction at room temperature stays within the hexagonal plane. The present results clearly show that the Al atom plays an important role in determining the easy magnetization direction in the \( Y_2Co_{15}Al_2 \) compound. The substitution of Al for Co leads to a decrease in the planar anisotropy and then changes the magnetic anisotropy from the basal plane to the c axis at room temperature. In an earlier study it was shown that the basal plane anisotropy has generally been attributed to the...
presence of the dumbbell pairs of Co atoms in the R$_2$Co$_{17}$ structure. Inomata$^2$ has performed point charge calculations at individual Co sites for the Y$_2$Co$_{17}$ compound and found that the Co atoms at 18f site contribution to the magnetocrystalline anisotropy is positive, whereas those at 6c, 9d, and 18h sites is negative. In particular, the 6c site is the largest negative contribution. This result agrees well with Streever’s nuclear magnetic resonance (NMR) studies in the R–Co intermetallic compounds.$^{41}$ Recently, a neutron powder diffraction study on the isotropic Nd$_2$Co$_{17-x}$Al$_x$ compounds has shown that the preference for Al atoms to occupy one of the four available Co sites in the rhombohedral Th$_2$Zn$_{17}$ structure decreases in the sequence 18h > 6c > 18f > 9d.$^{31}$ In fact, the 9d site is completely avoided by the Al atoms. Apparently the easy magnetization direction parallel to the c axis in the Y$_2$Co$_{15}$Al$_2$ compound is related to the preferential substitution of Al atoms on 6c and 18h sites.

Figure 7 shows the temperature dependence of magnetization measured in a field of 1 kOe at temperatures from 5 to 1000 K for the Pr$_2$Co$_{15}$Al$_2$ and Ho$_2$Co$_{15}$Al$_2$ compounds.

FIG. 7. Temperature dependence of the magnetization measured in a magnetic field of 1 kOe at temperatures from 1.5 to 1000 K for the Pr$_2$Co$_{15}$Al$_2$ and Ho$_2$Co$_{15}$Al$_2$ compounds.

$\alpha_J$ of Pr and Ho is negative and the anisotropy constant $K_1(R)$ is negative, which favors an easy magnetization direction perpendicular to the c axis. Therefore, the overall magnetocrystalline anisotropy of R$_2$Co$_{17}$ compounds with R=Pr and Ho is easy planar. However, in the substitution of Al for Co in R$_2$Co$_{17}$ the anisotropy of the Co sublattice may be changed from the planar anisotropy to the c-axis anisotropy. Thus, in R$_2$Co$_{15}$Al$_2$ compounds uniaxial anisotropy of the Co sublattice may overcome the planar anisotropy of the Pr and Ho sublattice, and a spin reorientation may appear.

In the case R$_2$Co$_{17}$ (where R=Sm, Er, and Tm), because $\alpha_J$ is positive for Sm, Er, Tm, a positive $K_1(R)$ is the result which is responsible for the easy c-axis magnetization. The uniaxial anisotropy of the R sublattice can overcome the planar anisotropy of the Co sublattice even at room temperature, leading to an easy c-axis anisotropy for R$_2$Co$_{17}$ compounds with R=Sm, Er, and Tm. The substitution of Al in R$_2$Co$_{17}$ leads to a positive $K_1(\text{Co})$ and also enhances the uniaxial anisotropy of these compounds. Therefore, a strong uniaxial anisotropy is expected for the Al-substituted R$_2$Co$_{17}$ compounds with R=Sm, Er, and Tm. We measured the magnetocrystalline anisotropy field of the Sm$_2$Co$_{15}$Al$_2$ compound at room temperature and found it to be 84 kOe, which is 13 kOe larger than that of Sm$_2$Co$_{17}$.\textsuperscript{42}

Our study demonstrates that the Ce$_2$Co$_{15}$Al$_2$ compound is uniaxial anisotropy over the whole magnetically ordered temperature range. Figure 8 shows the SPD results where the second derivative $d^2M/dH^2$ is plotted against the external field $H$ for the Ce$_2$Co$_{15}$Al$_2$ compound at room temperature.

FIG. 8. The SPD results where the second derivative $d^2M/dH^2$ is plotted against the external field $H$ for the Ce$_2$Co$_{15}$Al$_2$ compound at room temperature.

where the quantities in brackets are expectation. R$_2$Co$_{17}$ compounds have a negative $A_{20}$. The $\alpha_J$ of Pr and Ho is negative and the anisotropy constant $K_1(R)$ is negative, which favors an easy magnetization direction perpendicular to the c axis. Therefore, the overall magnetocrystalline anisotropy of R$_2$Co$_{17}$ compounds with R=Pr and Ho is easy planar. However, in the substitution of Al for Co in R$_2$Co$_{17}$ the anisotropy of the Co sublattice may be changed from the planar anisotropy to the c-axis anisotropy. Thus, in R$_2$Co$_{15}$Al$_2$ compounds uniaxial anisotropy of the Co sublattice may overcome the planar anisotropy of the Pr and Ho sublattice, and a spin reorientation may appear.

$K_1(R) = (-3/2)\alpha_J(r^2)(3J^2 - J(J + 1))A_{20}$, \textsuperscript{(3)}

where the quantities in brackets are expectation. R$_2$Co$_{17}$ compounds have a negative $A_{20}$. The $\alpha_J$ of Pr and Ho is negative and the anisotropy constant $K_1(R)$ is negative, which favors an easy magnetization direction perpendicular to the c axis. Therefore, the overall magnetocrystalline anisotropy of R$_2$Co$_{17}$ compounds with R=Pr and Ho is easy planar. However, in the substitution of Al for Co in R$_2$Co$_{17}$ the anisotropy of the Co sublattice may be changed from the planar anisotropy to the c-axis anisotropy. Thus, in R$_2$Co$_{15}$Al$_2$ compounds uniaxial anisotropy of the Co sublattice may overcome the planar anisotropy of the Pr and Ho sublattice, and a spin reorientation may appear.

In the case R$_2$Co$_{17}$ (where R=Sm, Er, and Tm), because $\alpha_J$ is positive for Sm, Er, Tm, a positive $K_1(R)$ is the result which is responsible for the easy c-axis magnetization. The uniaxial anisotropy of the R sublattice can overcome the planar anisotropy of the Co sublattice even at room temperature, leading to an easy c-axis anisotropy for R$_2$Co$_{17}$ compounds with R=Sm, Er, and Tm. The substitution of Al in R$_2$Co$_{17}$ leads to a positive $K_1(\text{Co})$ and also enhances the uniaxial anisotropy of these compounds. Therefore, a strong uniaxial anisotropy is expected for the Al-substituted R$_2$Co$_{17}$ compounds with R=Sm, Er, and Tm. We measured the magnetocrystalline anisotropy field of the Sm$_2$Co$_{15}$Al$_2$ compound at room temperature and found it to be 84 kOe, which is 13 kOe larger than that of Sm$_2$Co$_{17}$.\textsuperscript{42}

Our study demonstrates that the Ce$_2$Co$_{15}$Al$_2$ compound is uniaxial anisotropy over the whole magnetically ordered temperature range. Figure 8 shows the SPD results where the second derivative $d^2M/dH^2$ is plotted against the external field $H$ for the Ce$_2$Co$_{15}$Al$_2$ compound at room temperature. The value of $H_A$ is found to be 18 kOe, which is larger than that of Ce$_2$Co$_{17}$ [$H_A=7$ kOe at 300 K (Ref. 42)]. The enhancement of $H_A$ results from the increase of the c-axis anisotropy of the Co sublattice in Ce$_2$Co$_{15}$Al$_2$. However, it was found that the contribution to the bulk anisotropy from the Ce ions of Ce$_2$Co$_{17}$ is uniaxial. The c-axis anisotropy of Ce$_2$Co$_{17}$ was
confirmed by the measurement on the single crystalline and polycrystalline Ce$_2$Co$_{17}$. The hybridization of the Ce 4f states and the Co 3d states play an important role in determining the magnetic anisotropy of Ce$_2$Co$_{17}$. Therefore, it is apparent that the uniaxial anisotropy of the Ce$_2$Co$_{17}$Al$_2$ compound consists of two contributions originating from the Ce and Co sublattices.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China and has been carried out within the scientific exchange program between China and the Netherlands. Dr. Z. H. Cheng would like to thank the Alexander van Humboldt Foundation for financial support. The authors wish to express their gratitude to T. S. Ning and M. Hu for their assistance in works of magnetic measurement and experiment.