Magnetic Properties of $R_3Al_{11}(R=$La, Ce, Pr, Nd, Sm) Single Crystals

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Abstract. $R_3Al_{11}(R=$La, Ce, Pr, Nd) crystallizes in the orthorhombic La$_3$Al$_{11}$-type structure with two inequivalent $R$ sites. We have succeeded in growing these single crystals by using Al self flux method, and have performed the magnetization. All of the compounds except for nonmagnetic La$_3$Al$_{11}$ show successive phase transition and $R_3Al_{11}(R=$Ce, Pr, Nd) exhibits metamagnetism at low temperature. As reported previously, Ce$_3$Al$_{11}$ shows a ferromagnetic transition at 6.2 K and an antiferromagnetic one at 3.2 K for the $b$ axis and, in addition, we have found another anomalies inferred to be antiferromagnetic transition. Pr$_3$Al$_{11}$ shows an antiferromagnetic transition at 12.7 K and a ferromagnetic one at 3.5 K. Nd$_3$Al$_{11}$ shows three antiferromagnetic transitions at 13.5 K, 9.5 K and 2.9 K. Sm$_3$Al$_{11}$ does not follow the Curie-Weiss low and shows two antiferromagnetic transitions at 50 K and 26 K. These complex phase transitions and metamagnetism can be explained by two inequivalent $R$ ions with different crystal symmetry.

1. Introduction

Ce$_3$Al$_{11}$ crystallizes in to the orthorhombic La$_3$Al$_{11}$ type structure with the space group Immm. It has two inequivalent Ce ions sites with different crystal symmetry(CeI on the 2a site, CeII on the 4g site), and CeI and CeII show quite different behavior. Ce$_3$Al$_{11}$ shows a ferromagnetic transition at 6.2 K and an antiferromagnetic transition at 3.2 K where CeI orders magnetically and CeII owes Kondo scattering[1]. Additionally, it has a twin structure that have domain I with a lattice cell $(a$, $b$, $c)$ and another domain II with a lattice cell $(b$, $a$, $c)$ because of the relation $b \sim 3a$ between the lattice parameters. Recently, Ebihara et al have found that the antiferromagnetic order decrease rapidly with increasing with pressure but the ferromagnetic one does not by the measurement of electrical resistivity under pressure up to 3 GPa[2]. The purpose of this study is to clarify magnetic properties of two inequivalent Ce ions with different crystal symmetry for Ce$_3$Al$_{11}$ single crystals. For this purpose, it is important to study the magnetic property of $R_3Al_{11}(R=$La, Pr, Nd, Sm) single crystals. It is the first time as long as I know that the magnetic property of $R_3Al_{11}(R=$La, Pr, Nd, Sm) single crystal are reported.

2. Experiments

The single crystals were grown by using Al-rich self flux method. The starting composition of $R_7Al_{93}$ was placed in an Al$_2$O$_3$ crucible, sealed in a quartz ampoule and heated to 1000 °C and
then cooled to 750 °C at a rate of 15 °C/h and the Al flux was removed by centrifugation. The remaining flux was removed by 5 M NaOH solution. \( \text{R}_3\text{Al}_{11} (\text{R} = \text{La, Ce, Pr, Nd}) \) was square prism shape with dimension of \( \sim 1 \times 1 \times 6 \) mm\(^3\) and Sm\(_3\)Al\(_{11}\) was square plate shape with dimension of \( \sim 3 \times 3 \times 0.1 \) mm\(^3\). The purity and quality of the sample were checked by X-ray diffraction and Scanning Electron Microscopy/Energy Dispersion X-ray Spectrometry (SEM/EDS).

Magnetization measurements were carried out in a SQUID magnetometer (Quantum Design, MPMS-XL5) along the three axes \( a \), \( b \) and \( c \).

3. Results and Discussion

![Figure 1](image1.png)

**Figure 1.** (a) Isothermal magnetization curves for Ce\(_3\)Al\(_{11}\) single crystal. (b) Temperature dependence of the magnetization for Ce\(_3\)Al\(_{11}\) single crystal. Each symbol denotes as follows; circles: \( a \) axis. triangles: \( b \) axis. squares: \( c \) axis.

![Figure 2](image2.png)

**Figure 2.** Magnetic phase diagram of Ce\(_3\)Al\(_{11}\) single crystal. The open symbols obtained from M(T), and the closed symbols obtained from M(H).

Figure 1(a) shows isothermal magnetization curves for Ce\(_3\)Al\(_{11}\) single crystal at 1.8 K along the \( a \), \( b \) and \( c \) axis. Figure 1(b) shows temperature dependence of the magnetization for Ce\(_3\)Al\(_{11}\) single crystal in applied field of 0.01, 1.8, 2.5 T along the \( a \), \( b \) and \( c \) axis. Our result show almost the same result as reported by J. X. Boucherle et al[1]. In addition to that, we found another anomalies inferred to be antiferromagnetic transition along the \( a \) and \( b \) axis. Figure 2 shows magnetic phase diagram of Ce\(_3\)Al\(_{11}\) single crystal for three axes. We found three antiferromagnetic transition and one ferromagnetic transition.
Figure 3. (a) Isothermal magnetization curves for Pr$_3$Al$_{11}$ single crystal. (b) Temperature dependence of the magnetization for Pr$_3$Al$_{11}$ single crystal in a field of 0.1 T.

Figure 4. Magnetic phase diagram of Pr$_3$Al$_{11}$ single crystal.

Figure 3(a) shows isothermal magnetization curves for Pr$_3$Al$_{11}$ single crystal at 1.8 K along the $a$, $b$ and $c$ axis. Figure 3(b) shows the temperature dependence of the magnetization for Pr$_3$Al$_{11}$ single crystal in an applied field of 0.1 T along the $a$, $b$ and $c$ axis. They show an antiferromagnetic transition at 12.7 K and a ferromagnetic one at 3.5 K. Very sharp metamagnetic transition are induced by the magnetic field, involving a rearrangement of the moments for $a$ axis. Figure 4 shows Magnetic phase diagram of Pr$_3$Al$_{11}$ single crystal for three axes. We found two antiferromagnetic transitions and one ferromagnetic transition.

Figure 5(a) shows isothermal magnetization curves for Nd$_3$Al$_{11}$ single crystal at 1.8 K along the $a$, $b$ and $c$ axis. Figure 5(b) shows the temperature dependence of the magnetization for Nd$_3$Al$_{11}$ single crystal in an applied field of 0.1 T along the $a$, $b$ and $c$ axis. They show three antiferromagnetic transition at 13.5 K, 2.9 K and 9.5 K along the $a$, $b$ and $c$ axis respectively. Metamagnetic transition are induced by the magnetic field, involving a rearrangement of the moments for $b$ and $c$ axis. Metamagnetic transition is not observed for $a$ axis by the magnetization measurement field up to 5 T. Figure 6 shows Magnetic phase diagram of Nd$_3$Al$_{11}$ single crystal for three axes. We found three antiferromagnetic transitions.

Sm$_3$Al$_{11}$ shows two antiferromagnetic transitions at 50 K and 26 K along $a$ and $c$ axis respectively. As for the detail of this sample, We will report another paper.
Figure 5. (a) Isothermal magnetization curves for Nd$_3$Al$_{11}$ single crystal. (b) Temperature dependence of the magnetization for Nd$_3$Al$_{11}$ single crystal in a field of 0.1 T.

Figure 6. Magnetic phase diagram of Nd$_3$Al$_{11}$ single crystal.

4. Summary

Our results are similar to one separated the twinned domains by correction reported by Boucherle et al [1]. Single crystal growing by self flux method were not observed any twinned domains by magnetization measurements. The antiferromagnetic transition that has been observed for $b$ axis of Ce$_3$Al$_{11}$ was not observed $b$ axis of Pr$_3$Al$_{11}$. This suggests that the antiferromagnetic transition of $b$ axis in Ce$_3$Al$_{11}$ is invoked by Kondo effect of Ce ions. In the difference between magnitude of magnetization of Pr$_3$Al$_{11}$ after ferromagnetic transition and after metamagnetic transition, it may be causes that the Pr$_{I}$ site owe ferromagnetic order, and Pr$_{II}$ site owe antiferromagnetic order. It is expected that Nd$_3$Al$_{11}$ behavior is similar to Pr$_3$Al$_{11}$ one, and magnitude of magnetization of Nd$_3$Al$_{11}$ for $a$ axis after metamagnetic transition is larger than for $b$ axis one.

References

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