The negative volume magnetostriction of GdAl$_2$ with a cubic Laves structure

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Abstract. Spontaneous volume magnetostriction of the intermetallic compound GdAl$_2$ is studied from the X-ray diffraction measurement at low temperature. It was found that GdAl$_2$ exhibits the ferromagnetic transition at $T_C \sim 170$ K without lowering of crystal symmetry of a cubic C15 Laves structure. The temperature dependence of the lattice constant $a$ is in good agreement with that of the volume thermal expansion, in which negative spontaneous volume magnetostriction has been observed. It indicates that the magnetic exchange interaction can be described by the distance between the Gd moments. From these experimental results, the origin of the negative magnetostriction is discussed on the basis of the localized spin model of ferromagnetism.

1. Introduction

The magnetostriction is a general effect in solid state physics because any magnetization is directly connected to a lattice deformation. Generally, ferromagnetic Inver alloys such as Fe-Ni and Fe-Pt show very strong mutual dependence between magnetism and volume. They exhibit a very small or negative thermal expansion coefficient below the ferromagnetic temperature $T_C$ and show the large spontaneous volume magnetostriction of $\omega \sim 10^{-2}$[1], which is defined as a relative volume change accompanying the phase transition from the high-temperature paramagnetic state to the low-temperature state with a spontaneous magnetic order. Such a large magnetovolume effect has been investigated on the basis of the itinerant electron model of ferromagnetism[2]. On the other hand, some rare earth compounds show the negative spontaneous volume magnetostriction $\omega$, which cannot be explained in terms of the theory based on general spin fluctuations[2]. It may comes from the difference between the origin of the magnetism of the localized 4f electron and that of itinerant electrons in the 3d band couple ferromagnetically.

Here we focus the magnetovolume effects of RAl$_2$ (R: Rare earth element) compounds which crystallizes C15 cubic Laves structure. GdAl$_2$ has the highest $T_C(\sim 170$ K) among these ferromagnetic compounds. The previous reports have revealed that GdAl$_2$ has 5d-like electrons[3] and its spontaneous magnetic moment lies between 6.83 and 7.20 $\mu_B$[4]. The magnetovolume effect is anomalous because the negative spontaneous volume magnetostriction is obtained to be $\omega \sim -10^{-3}$[5]. In the present work, the magnetic susceptibility and the X-ray diffraction...
experiments were carried out through $T_C$ for GdAl$_2$ in order to make clear the magnetovolume effect and the stability of crystal structure at the ferromagnetic state.

2. Experimental

Single crystals were grown by a Czochralski pulling method[6, 7]. The pulling parameter was kept constant during the growth (pulling rate: 10 mm/h; seed rotation speed: 10 rpm; crucible-rotation speed: 5 rpm). An ingot was 3-4 mm in diameter and 50 mm in length. The single crystalline state was confirmed using back-scattering Laue technique. The DC magnetic susceptibility was measured at ambient pressure using a Quantum Design MPMS-5 superconducting quantum interference device magnetometer.

A powder sample was obtained by grinding the single crystal. X-ray powder diffraction measurements were performed at low temperature using the RINT 2500 system, Rigaku Co, with a graphite counter monochromator and an X-ray generator with a rotating Cu anode. The generator was operated at 50 kV and 300 mA. The sample was fixed in a closed-cycle He gas refrigerator mounted on a diffractometer[8]. At several temperatures entire profiles of reflection peaks were measured with a step size of 0.01° and the step-counting time of 6 s. From the observed profile the lattice constant and the full width at half maximum (FWHM) were obtained.

3. Result and discussion

Figure 1 shows the temperature dependence of the DC magnetic susceptibilities and the inverse magnetic susceptibility of GdAl$_2$, in which magnetic field of 1 kOe is applied along the (100), (110), and (111) axes. At the paramagnetic phase above $T_C$, magnetic susceptibilities follow the Curie-Weiss law and are isotropic at the magnetic field for all axes because of the vanishing orbital moment ($L=0$) of the Gd ion. The effective magnetic moment per Gd atom are 8.9$\mu_B$, 9.0$\mu_B$ and 9.1$\mu_B$ along (100), (110) and (111) axes, respectively, which are larger than that of the free Gd$^{3+}$ ion value, 8.0$\mu_B$. The paramagnetic Curie temperatures are 173.8 K, 173.1 K and 172.1 K along (100), (110) and (111) axes, respectively. At the ferromagnetic state below $T_C$, on the other hand, $\chi(T)$ along the (110) axis is much larger than that for the other axes, indicating that the easy magnetic direction is parallel to the (110) axis.

![Figure 1](image-url). Magnetic susceptibility at the magnetic field along several axes (solid symbols) and the inverse magnetic susceptibility (open circles) of GdAl$_2$. The dashed line is the Curie-Weiss law.
Figure 2. Observed X-ray profiles of GdAl$_2$ at 12 K. The asterisk shows a impurity peak.

Figure 2 shows the observed X-ray diffraction profile at 12 K. It can be seen that all the peaks in the diffraction patterns can be indexed to the cubic C15 Laves phase ($Fd\bar{3}m$), which is same as that at room temperature. In figure 3, a FWHM of (311) reflection is plotted against temperature. Inset shows the profile at 12 K in the expanded scale. Although the FWHM increases slightly with decreasing temperature, a clear split of the diffraction spectrum due to the ferromagnetic transition was not observed. It means that GdAl$_2$ exhibits the ferromagnetic transition without lowering of crystal symmetry as well as a relative volume changes isotropically.

Based on the results of the profile, the temperature dependence of the lattice constant $a$ of GdAl$_2$ are obtained as shown in Figure 4. Although the value of $a$ decreases almost linearly with decreasing temperature from 300 K, the slope $da/dT$ below $T_C$ becomes steeper than that above $T_C$. For comparison, the value of $a$ was calculated from the temperature dependence of the volume thermal expansion in the previous result[5] and plotted as the solid line in Figure 4. The obtained lattice parameter $a$ from the volume thermal expansion is in good agreement with that obtained in the present work.

On the basis of the itinerant electron model of ferromagnetism, it is impossible to explain the negative volume magnetostriction below $T_C$[2]. A possible scenario is that the local magnetic moments and equilibrium lattice parameters are evaluated for two spin structures, for the ferromagnetic ground state and for a state with randomly oriented Gd moments; that is, 4$f$ electrons are well localized at each Gd ion of GdAl$_2$ and the negative sign of the magnetostriction comes from the decrease of the total energy difference between the paramagnetic state and the ferromagnetic one with increasing volume. Since GdAl$_2$ exhibits the ferromagnetic transition without lowering of crystal symmetry, we can assume the simple exchange interactions in GdAl$_2$ in terms of an effective classical Heisenberg Hamiltonian, where the magnetic exchange interaction can be described by the distance between the local magnetic moments in each Gd ion[9].
4. Summary

Magnetic susceptibility measurement and X-ray diffraction measurement has been carried out for GdAl$_2$ which shows ferromagnetic transition at $T_C \sim 170$ K. Above $T_C$, magnetic susceptibilities are isotropic at the magnetic field for all axes and follow the Curie-Weiss law. The negative volume magnetostriction of GdAl$_2$ is clearly observed from the results of the X-ray diffraction measurement as well as that of the volume thermal expansion. On the other hand, no structural transition has been observed at $T_C$ keeping a cubic C15 Laves structure. It indicates that the magnetic exchange interaction can be described by the distance between the Gd moments. From these experimental results, the origin of the negative magnetostriction is discussed on the basis of the localized spin model of ferromagnetism.

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