Magnetic properties of Nd$_x$Er$_{1-x}$Mn$_6$Sn$_6$ alloys

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Abstract. We have conducted low- and high-field magnetization measurements and neutron diffraction experiments on Nd$_x$Er$_{1-x}$Mn$_6$Sn$_6$ alloys (0 $\leq$ x $\leq$ 0.5) with hexagonal MgFe$_6$Ge$_6$ type structure. The alloy with x = 0.1 shows a complex helical antiferromagnetism with the magnetic moments rotating in the c plane below the Curie temperature $T_C$ = 340 K, and a ferrimagnetism below the temperature $T_I$ = 100 K. In the ferrimagnetic state, the magnetic moments $\mu_{\text{Mn}}$ and $\mu_{\text{Nd}}$ are parallel to each other, and antiparallel to $\mu_{\text{Er}}$. The alloy with x = 0.4 shows also a similar ferrimagnetic structure below the Curie temperature $T_C$ = 360 K. It is likely that the each magnetic moments are a keep constant values of $\mu_{\text{Mn}}$ = 2.3 $\mu_B$/Mn atom, $\mu_{\text{Nd}}$ = 3 $\mu_B$/Nd atom and $\mu_{\text{Er}}$ = 9 $\mu_B$/Nd atom at 10 K in the whole composition (0 $< x \leq$ 0.5). The ferrimagnetic arrangements for the alloys with 0.2 $\leq x \leq$ 0.5 are not almost disturbed by magnetic field up to 26 T at 4.2 K.

1. Introduction

Ternary RMn$_6$Sn$_6$ (R = Y, Tb, Dy and Er) alloys have the hp13 type (MgFe$_6$Ge$_6$ type) layer structure. It should be noted that Mn atom layers on 6i ($\frac{1}{2}$, 0, z$_2$) and ($\frac{1}{2}$, 0, $\pm$z$_1$) are well separated by Sn atom layers on 2c ($\frac{1}{2}$, $\frac{3}{2}$, 0) and 2e (0, 0, $\pm$z$_1$), and by Sn atom layers on 2d ($\frac{1}{2}$, $\frac{3}{2}$, $\frac{1}{2}$) and R atom layer on 1a (0, 0, $\frac{1}{2}$), where z$_1$ and z$_2$ are nearly equal to 0.16 ($\sim$ $\frac{1}{6}$) and 0.25 ($\sim$ $\frac{1}{3}$), respectively. The YMn$_6$Sn$_6$ alloy shows an inhomogeneous helical antiferromagnetism with the Néel temperature $T_N$ = 333 K and the paramagnetic Curie temperature $\theta_P$ = 394 K [1, 2]. We have made magnetization measurements for the YMn$_6$Sn$_6$ alloy, and obtained interesting results; the magnetization approaches saturation around 11 T at 77 K, and the saturated magnetization corresponds to the magnetic moment of 2.0 $\mu_B$/Mn atom [3]. The isotypic Tb$_x$Y$_{1-x}$Mn$_6$Sn$_6$ alloy with x = 0.2 shows a helical arrangement with propagation vector $\mathbf{q}$ = (0.133 0 0) at 10 K [3]. Recently, we have reported that the isotypic Dy$_x$Y$_{1-x}$Mn$_6$Sn$_6$ alloy with x = 0.5 shows a ferrimagnetic structure which the magnetic moments of Mn, $\mu_{\text{Mn}}$ = (2.3 $\mu_B$/Mn atom), are parallel to each other, and antiparallel to the magnetic moments of Dy atom, $\mu_{\text{Dy}}$ = (10 $\mu_B$/Dy atom) [4], as similar to the magnetic structure of the DyMn$_6$Sn$_6$ alloy at low temperature [5]. More recently, we have also reported the isotypic Nd$_x$Y$_{1-x}$Mn$_6$Sn$_6$ alloy with x = 0.2 shows a helical antiferromagnetism with propagation vector $\mathbf{q}$ = (0, 0, $\frac{1}{2}$) at 10 K [6]. On the other hand, the ErMn$_6$Sn$_6$ alloy shows a complex helical antiferromagnetism with the Néel temperature $T_N$ = 352 K, and a ferrimagnetsim below the temperature $T_I$ = 75 K [5]. It should be noted that a metamagnetic transition is observed for the ErMn$_6$Sn$_6$ alloy in a field higher than around 20 T at 4.2 K [7]. We have found that Nd$_x$Er$_{1-x}$Mn$_6$Sn$_6$ alloys have the hp13-type structure in the composition range of 0 $\leq$ x $\leq$ 0.5. The present paper reports on
the results of low- and high-field magnetization measurements, X-ray and neutron diffraction experiments for the pseudo ternary Nd$_{x}$Er$_{1-x}$Mn$_6$Sn$_6$ alloy system.

2. Sample preparation and experimental procedure
The method of sample preparation was described in our earlier paper [3]. Magnetizations were measured by a vibrating sample magnetometer (VSM) in a field up to 2 T, a VSM up to 13 T at IMR and an extraction method up to 26 T produced by a hybrid magnet 28T-HM at the High Field Laboratory of Tohoku University. X-ray diffraction experiments were made using a conventional diffractometer with a copper target. Neutron diffraction experiments were made for powder samples using HERMES of IMR installed in the JRR-3M reactor at JAERI.

3. Experimental Results and Discussion
The lattice constants $a$ and $c$ increase slightly with increasing $x$; for example, $a = 5.516$ Å and $c = 8.999$ Å for $x = 0.0$ and $a = 5.541$ Å and $c = 9.019$ Å for $x = 0.5$ at 293 K.

Figure 1 shows the magnetization versus temperature curves in various fields up to 2.0 T for the Nd$_{0.1}$Er$_{0.9}$Mn$_6$Sn$_6$ alloy. The curve in a field of $1.0 \times 10^{-2}$ T shows anomalies at temperatures $T_t (= 100 \text{ K})$ and $T_N (= 340 \text{ K})$ as shown by the arrows in Fig. 1. We assumed that $T_t$ and $T_N$ are magnetic structure changes from a ferrimagnetism to a helical antiferromagnetism and the Néel temperature, respectively. The transition temperature $T_t$ increases with increasing field up to 0.6 T. It is likely that a ferrimagnetic arrangement only appears in fields higher than 1.0 T below the Curie temperature $T_C$. Figure 2 shows the results for the Nd$_{0.4}$Er$_{0.6}$Mn$_6$Sn$_6$ alloy. We also determined $T_C (= 360 \text{ K})$ from the curve in a field of $1.0 \times 10^{-2}$ T. The temperatures of $T_N$ or $T_C$ slightly increase with increasing $x$ from 352 K for $x = 0.0$ to 364 K for $x = 0.5$.

Figure 3 shows neutron diffraction pattern for the Nd$_{0.1}$Er$_{0.9}$Mn$_6$Sn$_6$ alloy at 10 K. There are strong magnetic 001 reflection in addition to the nuclear reflections. Since Nd concentration in this alloy was poor, we assumed a fictional atom with average neutron scattering amplitude of Nd and Er atoms on the 1a site and the magnetic moment of its atom, $\mu_{1a}$, with average magnetic moment of Nd and Er atoms, $\mu_{Nd}$ and $\mu_{Er}$, and performed an analysis. The results are as follows; the hexagonal lattice constants are $a = 5.487$ Å and $c = 8.963$ Å, the 2c, 2d and 2e sites are entirely occupied by Sn atoms, the 6i site is occupied by Mn atoms, and the 1a site is randomly occupied by Nd and Er atoms, the magnetic moments of Mn atom $\mu_{Mn}(= 2.3 \mu_B / \text{Mn atom})$ and $\mu_{1a}(= 8 \mu_B / \text{Nd or Er atom})$ lay in the $c$ plane, and are antiparallel to each other.
Figure 3. Neutron diffraction pattern for Nd$_{0.1}$Er$_{0.9}$Mn$_6$Sn$_6$ alloy at 10 K.

Figure 4. Neutron diffraction pattern for Nd$_{0.1}$Er$_{0.9}$Mn$_6$Sn$_6$ alloy at 130 K.

Figure 5. Neutron diffraction pattern for Nd$_{0.4}$Er$_{0.6}$Mn$_6$Sn$_6$ alloy at 10 K.

Figure 4 also shows neutron diffraction pattern for the Nd$_{0.1}$Er$_{0.9}$Mn$_6$Sn$_6$ alloy at 130 K. It should be noted that a satellite reflection, 001-δ, appears, but the 001 and 001+δ reflections are almost absent in the pattern, suggesting a complex helical magnetic structure. A detailed analysis is now in progress. Figure 5 shows neutron diffraction pattern for the Nd$_{0.4}$Er$_{0.6}$Mn$_6$Sn$_6$ alloy at 10 K. This pattern is similar to the one for the alloy with $x = 0.1$ at 10 K. We also determined that the Mn, Nd and Er atoms occupy on the ordinary sites as case of the alloy with $x = 0.1$, and the magnetic moments, $\mu_{\text{Mn}} = 2.3\, \mu_B$/Mn atom and $\mu_{\text{1a}} = 4\, \mu_B$/Nd or Er atom, lay in the c plane, and are antiparallel to each other.

If Nd and Er atoms obey Hund’s rule, and $\mu_{\text{Nd}}$ and $\mu_{\text{Er}}$ are antiparallel to each other, we obtained the calculated magnetic moments $\mu_{\text{Nd}}^c = 3.27\, \mu_B$/Nd atom, $\mu_{\text{Er}}^c = 9.0\, \mu_B$/Er atom, $\mu_{\text{1a}}^c = 7.87\, \mu_B$/Nd or Er atom for alloy with $x = 0.1$, and $\mu_{\text{1a}}^c = 4.08\, \mu_B$/Nd or Er atom for alloy with $x = 0.4$. The values of calculated and observed magnetic moments, $\mu_{\text{1a}}^c$ and $\mu_{\text{1a}}$, are not so different in respective alloys. So we conclude that the magnetic moments of Mn, Nd and Er atoms keep constant values of $\mu_{\text{Mn}} = 2.3\, \mu_B$/Mn atom, $\mu_{\text{Nd}} = 3\, \mu_B$/Nd atom and $\mu_{\text{Er}} = 9\, \mu_B$/Er atom in the whole composition $0 < x \leq 0.5$ at 10 K. On the other hand, the high field magnetization measurements for the ErMn$_6$Sn$_6$ alloy using a single crystal have been made by T. Suga et al; in part of the lower field, the magnetization saturates around $4\, \mu_B$/f.u. when the magnetic field is applied parallel to the b axis [7]. If the saturation magnetization is $\mu_F = 4\, \mu_B$/f.u. and $\mu_{\text{Er}} = 9\, \mu_B$/Er atom, we can estimate $\mu_{\text{Mn}} = 2.17\, \mu_B$/Mn atom.

Figure 6 shows the field dependence of the magnetic moment $\mu$ (f.u.) for the Nd$_x$Er$_{1-x}$Mn$_6$Sn$_6$ alloys with $x = 0.1$, 0.3 and 0.5 obtained by using a VSM in fields up to 13 T at 4.2 K at IMR. Magnetization reaches saturation in a field of several Tesla. After saturation, magnetization slightly increases with the increasing field. The magnetizations in a field of 13 T are also shown.
in Fig. 1 and 2 as closed circles at 4.2 K and 77 K. The magnetization in a high-field increases with increasing temperature; this suggests that the magnetic moment $\mu_{Er}$ rapidly decreases with increasing temperature in ferrimagnetic structure. The concentration $x$ dependence of the magnetization $\mu$ (f.u.) in a field of 13 T is also shown in Fig. 7 as closed circle. The magnetizations in a field of 13 T linearly increase with increasing $x$. We have also made high-field magnetization measurements up to a field of 26 T using a hybrid magnet 28T-HM. After saturation, the magnetization slightly increases with increasing magnetic fields up to a field of 26 T at 4.2 K. The magnetizations in a field of 26 T are also shown in Fig. 7 as symbol ⊙. We observed no such metamagnetic transition as seen in the ErMn$_6$Sn$_6$ alloy. It can be concluded that a metamagnetic transition field increases with increasing $x$.

4. Conclusion
We have concluded that the Nd$_x$Er$_{1-x}$Mn$_6$Sn$_6$ alloys (0 $\leq$ $x$ $\leq$ 0.5) show a ferrimagnetic structure at low temperature. The magnetic moments $\mu_{Mn}$, $\mu_{Nd}$ and $\mu_{Er}$ lay in the $c$ plane, $\mu_{Mn}$ and $\mu_{Nd}$ are parallel to each other, and antiparallel to $\mu_{Er}$, and $\mu_{Mn}$= 2.3$\mu_B$/Mn atom), $\mu_{Nd}$= 3$\mu_B$/Nd atom) and $\mu_{Er}$= 9$\mu_B$/Er atom) are kept at constant values in the whole composition at 10 K. We also concluded that the ferrimagnetic arrangements for the alloys with 0.2 $\leq$ $x$ $\leq$ 0.5 are kept in a magnetic field up to 26 T at 4.2 K.

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