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MAGNETIC PROPERTIES OF PSEUDO-BINARY COMPOUNDS \((\text{Nd}_{1-x}\text{Lu}_x)\text{Mn}_2\) AND \((\text{Gd}_{1-x}\text{Lu}_x)\text{Mn}_2\)

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Abstract. It has been found that a large Mn moment in \(\text{RMn}_2\) (\(R = \text{Nd and Gd}\)) disappears with decreasing the lattice constant by substituting \(R\) for \(\text{Lu}\). From the results of magnetic and NMR measurements on \((\text{Gd}_{1-x}\text{Lu}_x)\text{Mn}_2\), the magnetic spin structure and the magnitude of Mn moments are deduced.

It has been pointed out that the onset of Mn moment in Laves phase compounds \(\text{RMn}_2\) (\(R = \text{rare earths and Y}\)) correlates with the Mn-Mn distance, \(d_{\text{Mn}-\text{Mn}}\) [1, 2]. Namely, the Mn sublattice in \(\text{RMn}_2\) has a large moment for \(R = \text{Y, Pr, Nd, Sm, Gd and Tb}\) with relatively large \(d_{\text{Mn}-\text{Mn}}\), while it has nearly zero moment for \(R = \text{Dy, Ho, Er, Tm and Lu}\) with small \(d_{\text{Mn}-\text{Mn}}\) at low temperature. In this paper, we present the results of magnetization, thermal expansion and NMR measurements on \((\text{R}_{1-x}\text{Lu}_x)\text{Mn}_2\) with \(R = \text{Nd and Gd}\), where \(\text{Lu}\) is a nonmagnetic atom with the smallest atomic radius in lanthanide.

The samples were prepared by a plasma-jet melting, followed by annealing at 700-850 °C for a week. The powder X-ray diffraction at room temperature shows that all the \((\text{Nd}_{1-x}\text{Lu}_x)\text{Mn}_2\) crystallize into the C14 structure, while \((\text{Gd}_{1-x}\text{Lu}_x)\text{Mn}_2\) into the C15 one for \(x < 0.3\) and the C14 one for \(0.5 < x < 1.0\), where the lattice constant of each system decreases continuously with increasing \(x\).

The results obtained are summarized in figures 1 and 2.

Fig. 1. (a) Susceptibility at \(H = 3.0\) kOe and (b) thermal expansion \(\Delta l/l\) as a function of temperature for \((\text{Nd}_{1-x}\text{Lu}_x)\text{Mn}_2\). The \(\Delta l/l\) was measured by a dilatometer.

Fig. 2. (a) Thermal expansion curves measured by a dilatometer and (b) \(^{55}\text{Mn}\) spin-echo NMR spectra observed under zero field at 4.2 K for \((\text{Gd}_{1-x}\text{Lu}_x)\text{Mn}_2\).
has no distinct anomaly against temperature. These confirm that LuMn$_2$ is a Pauli paramagnet as reported before [4].

In the Gd system, $T_N$ decreases monotonously with increasing $x$ as well as in the Nd system (see Fig. 2a). The reduction of $T_N$ in both the Nd and Gd systems suggests that the antiferromagnetic interaction between Mn moments is weakened by shrinking the lattice. The $\omega_s$ at $T_N$ is found to be almost constant for $0 \leq x \leq 0.2$, but it decreases suddenly beyond $x = 0.3$ and disappears for $x \geq 0.5$ with smaller $d_{\text{Mn-Mn}}$ than that of 2.64 Å at 4.2 K. The zero field $^{55}$Mn NMR spectra at 4.2 K for $0 \leq x \leq 0.3$, all of which crystallize into the C15 cubic structure, are shown in figure 2b. The NMR signals are observable around 120 MHz in (Gd$_{1-x}$Lu$_x$)Mn$_2$ with $x \leq 0.2$, and other weak signals appear around 30 MHz in addition to 100 MHz for $x = 0.3$. It has been found that the integrated intensities of the spectra around 100-120 MHz decrease with increasing $x$. Since the signals distributed around 100-120 MHz and 30 MHz seem to be due to the Mn atoms having a large moment and no moment, respectively [1], these results suggest that the number of Mn atoms with a large moment decreases, while that with no moment increases with increasing $x$. It is worth to note that $\omega_s$ at $T_N$ is almost constant for $x \leq 0.2$, while a large number of Mn atoms lose their large moments with increasing $x$. This might be interpreted to mean that the magnitude of reduction of the amplitude of spin fluctuation $\langle \mu_{\text{Mn}}^2 \rangle$ at $T_N$ increases with the increase of $x$, because $\omega_s$ is proportional to $\langle \mu_{\text{Mn}}^2 \rangle$ suggested by Shiga [5].

As is evident from figure 3a, the (Gd$_{1-x}$Lu$_x$)Mn$_2$ compounds with $x \leq 0.3$ have a large high field susceptibility, $\chi_M$, of $\sim 3 \times 10^{-4}$ emu/g, while $\chi_M$ tends to nearly zero for $x \geq 0.5$. This implies that a drastic change on magnetic structure occurs around $x = 0.4$.

In the previous paper [6], we proposed that a Gd-canted and Mn antiferromagnetic (GAMA) structure or a Gd antiferromagnetic and Mn antiferromagnetic (GAMA) one is realized at low temperature in GdMn$_2$. It seems likely that GAMA model for $0 \leq x \leq 0.2$ and GcMA one for $x = 0.3$, which are shown in figure 3b, are reasonable, because the initial slope in the magnetization vs. applied field curve for $x \leq 0.2$ is clearly smaller than that for $x = 0.3$ on spherical samples prepared carefully. The GCMA structure could be developed for $x \leq 0.2$ as a meta-stable state when the field is applied. The Mn moments for $x \leq 0.3$ have been estimated by assuming that the hyperfine coupling constant of 42.7 kOe/µ$_G$ in the antiferromagnetic YMn$_2$ [7] is applicable in this system. The transfer hyperfine field from the Gd sublattice can be neglected for $x \leq 0.2$ because of the antiferromagnetic configuration in Gd sublattice. A collinear ferrimagnetism seems to be established for $x \geq 0.5$, where the Mn moments induced by the exchange field from the ferromagnetic Gd sublattice are coupled antiferromagnetically with the Gd moments as in GdCo$_2$. Assuming that the Gd atoms have a magnetic moment of 7.0 µ$_G$ expected from Gd$^{3+}$ free ion, the Mn moments for $x \geq 0.5$ are estimated. The results are shown in figure 3b. It is seen that the magnitude of Mn moment decreases rapidly around $x = 0.4$, where the $d_{\text{Mn-Mn}}$ is very close to a critical distance of 2.67 Å at 4.2 K.

From these results, it can be concluded that the Mn atoms in (R$_{1-x}$Lu$_x$)Mn$_2$ with R = Nd and Gd lose their ground-state large moments in the compounds which have smaller $d_{\text{Mn-Mn}}$ than a critical value of about 2.67 Å at 4.2 K. The spin configuration in rare earth sublattice changes with the drastic variation of Mn sublattice moments.