

## First-Principles Study of $R\text{Fe}_{12}$ -type Magnet Compounds

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$R\text{Fe}_{12}$  compounds ( $R$  being rare-earth element) with the  $\text{ThMn}_{12}$  structure have been attracting interest as possible strong permanent magnet compounds. They contain the highest iron content among known rare-earth iron intermetallics, which is favorable for achieving large saturation magnetization. Recently,  $\text{NdFe}_{12}\text{N}$  has been synthesized as a film on top of W underlayer on MgO substrate, and it was shown to have larger saturation magnetization and anisotropy field than  $\text{Nd}_2\text{Fe}_{14}\text{B}$  [1]. However,  $R\text{Fe}_{12}$  and  $R\text{Fe}_{12}\text{N}$  are thermodynamically unstable, and it is necessary to substitute a part of Fe atoms with another element for bulk stability.

In the present work, we study theoretically the magnetism and structural stability of  $R\text{Fe}_{12}$  and  $R\text{Fe}_{12}X$  ( $X=\text{B}, \text{C}, \text{N}, \text{O}, \text{F}$ ), and discuss the role of substitution element and  $X$  element. The calculation is based on the density functional theory in the generalized gradient approximation (GGA). The  $R$ -4f electrons are treated as open-core. We use QMAS code [2] based on the projector augmented wave (PAW) method with the planewave basis set.

Figure 1 shows the magnetic moment and magnetization (per volume) of  $\text{NdFe}_{11}\text{Ti}X$  with  $X=\text{B}, \text{C}, \text{N}, \text{O}, \text{F}$  [3]. The magnetization is enhanced compared to  $\text{NdFe}_{11}\text{Ti}$  for all  $X$  by the magnetovolume effect. We also see that the magnetic moment varies with changing  $X$ , and the cases with  $X=\text{N}, \text{O}, \text{F}$  have larger magnetic moment than the  $X=\text{B}, \text{C}$  cases. Similar  $X$  dependence has been reported in other iron compounds as well [4,5]. In the case of  $X=\text{B}$ , the magnetic moment is suppressed at the Fe(8j) site, whereas it is enhanced at the Fe(8f) site. This change is explained by *cobaltization*. As the atomic number of  $X$  increases, the  $X$ -2p state is pulled down, and eventually the anti-bonding states between  $X$ -2p and Fe-3d are partially occupied in the majority spin channel. This results in jump of the magnetic moment between  $X=\text{C}$  and  $X=\text{N}$ . On the other hand, the second-order crystal-field parameter takes a maximum at  $X=\text{N}$ . These results suggest that nitrogen is the best dopant among B, C, N, O, and F in terms of magnetization and magnetocrystalline anisotropy. We also computed intersite magnetic exchange coupling in  $\text{NdFe}_{12}X$  by the Liechtenstein method using Akai-KKR code [6]. Recent spin model analyses clarified that the Nd-Fe coupling is essential for magnetocrystalline anisotropy at high temperature [7,8]. Our first-principles calculation shows that the Nd-Fe couplings are strongly affected by the presence of  $X$ . Especially, the Nd-Fe(8j) coupling is reduced significantly by  $X=\text{N}$ . We will discuss electronic origin of this change in more detail.

We also study  $\text{NdFe}_{11}M$  for  $M=\text{Ti}, \text{V}, \text{Cr}, \text{Mn}, \text{Co}, \text{Ni}, \text{Cu}, \text{Zn}$  [9]. The formation energy is negative in  $M=\text{Ti}$ . This suggests that Ti stabilizes the system, which is consistent with experimental observation. However, Ti leads to substantial reduction of magnetic moment. In the case of  $M=\text{Co}$ , on the other hand, the formation energy is negative and magnetic moment is comparable to that of  $\text{NdFe}_{12}$ . Therefore, cobalt could be a good stabilizing element for  $\text{NdFe}_{12}X$ .

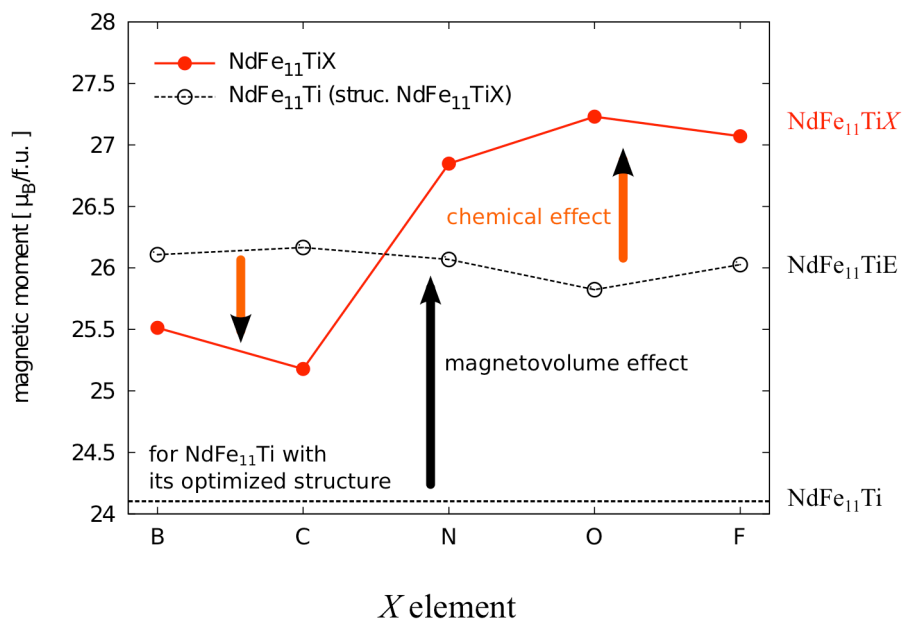


Fig. 1: Magnetization of  $NdFe_{11}TiX$  ( $X=B, C, N, O, F$ ). The open circles show the results for  $NdFe_{11}Ti$  with the optimized structure for  $NdFe_{11}TiX$  [3].

#### References

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