Ab initio inspection on doping effects on rare-earth permanent magnets

Munehisa Matsumoto (Akai Group)
Institute for Solid State Physics, University of Tokyo
Kashiwa-no-ha, Kashiwa, Chiba 277-8581

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We have been exploring possibilities to improve rare-earth permanent magnets (REPM's) on the basis of fundamental understanding for the intrinsic electronic structure. The champion magnet, Nd-Fe-B ternary alloy, is made of the main-phase compound $Nd_2Fe_{14}B$. Its excellent magnetic properties at room temperature are traded off with a drawback with poor high-temperature performance. The possible directions for improving the intrinsic properties of REPM's include raising the Curie temperature and/or enhancing the temperature resistance of magnetic properties.

A standard way to raise the Curie temperature is to make an alloy with Co. Thus it is important to understand how Co-doping works in $Nd_2Fe_{14}B$. Even though some experimental observation on the site preference of doped elements in $Nd_2Fe_{14}B$, it is only in recent days that *ab initio* investigations on $Nd_2Fe_{14}B$ have been in major trends. This is partly because the large unit cell of $Nd_2Fe_{14}B$ with 4 formula units or 68 atoms is demanding in terms of computational resource. Also this practical problem comes on top of the fundamental problem in dealing with $4f$ electrons. At the core of our calculations are open-source package for *ab initio* electronic structure calculations, AkaiKKR [1] and OpenMX [2]. With these the instrinsic properties of doped $Nd_2Fe_{14}B$ can be addressed in a reasonable time on System B. Continuous exploration of the compositinal space is done on the basis of coherent potential approximation using AkaiKKR. Effect of discrete replacement of host atoms by dopant atoms is investigated by OpenMX. Part of our recent results on Co-doped $Nd_2Fe_{14}B$ where one Co atoms replaces one Fe atom out of the possible 56 atomic sites in the unit cell of $Nd_2Fe_{14}B$ is shown in Fig. 1 for the mixing energy which is defined as follows.

$$\begin{aligned}\Delta E_{\rm mix} &= U[{\rm Nd_2(Fe_{1-\mathit{x}}Co_{\mathit{x}})_{14}B}] \\ &\quad -(1-x)U[{\rm Nd_2Fe_{14}B}] \\ &\quad -xU[{\rm Nd_2Co_{14}B}]\end{aligned}$$

Here $U[M]$ is the calculated energy for a given material M. It is clearly shown that doped

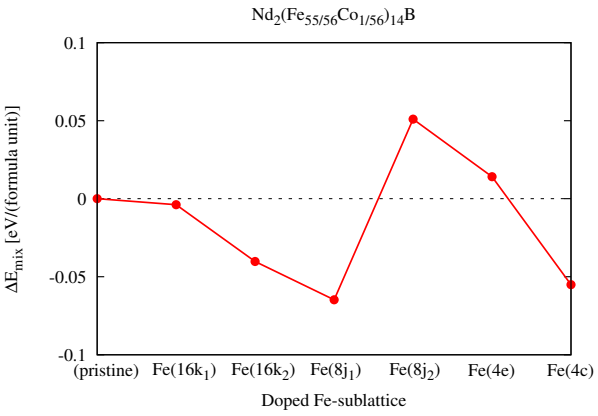

Figure 1: Calculated mixing energy of Co and $Nd_2Fe_{14}B$.

Co prefers Fe($8j_1$) sublattice among the six

sublattices in the host $Nd_2Fe_{14}B$ [3]. Remarkably the numerically observed trend is consistent with past experimental claims in that Co prefers Fe($8j_1$) and avoids Fe($8j_2$). It also can be shown that this particular preference originates in the magnetic exchange couplings between the host Fe and dopant atoms. Such fundamental understanding should pave the way toward the improvement of intrinsic properties avoiding unnecessary tradeoff's between various desired properties to make a good REPM.

References

[1] H. Akai: `http://kkr.issp.u-tokyo.ac.jp/jp/`. See also `https://ma.issp.u-tokyo.ac.jp/app/113`.

[2] T. Ozaki *et al*: `http://www.openmx-square.org/`. See also `https://ma.issp.u-tokyo.ac.jp/app/594`.

[3] M. Matsumoto, preprint (arXiv: 1812.10945)