Curie temperature of Sm$_2$Fe$_{17}$ and Nd$_2$Fe$_{14}$B: a first-principles study

Taro Fukazawa (AIST, ESICMM NIMS)
Hisazumi Akai (ISSP, ESICMM NIMS)
Yosuke Harashima (AIST, ESICMM NIMS)
Takashi Miyake (AIST, ESICMM NIMS)

In his development of the Nd-Fe-B magnet, Sagawa had a working hypothesis: If the shortest Fe-Fe bond in R$_2$Fe$_{17}$ (R: rare-earth element) could be elongated, its ferromagnetic state would become more stable. [1] Although he used boron with the intention of stretching the bond, the resulting crystal, Nd$_2$Fe$_{14}$B, has utterly different structure from R$_2$Fe$_{17}$. In the present paper, we address two questions related to this episode—whether the hypothesis is valid and how boron affects the Curie temperature ($T_C$) in Nd$_2$Fe$_{14}$B.

The hypothesis is seemingly supported by later works. Li and Morrish deduced a negative exchange for the bond, and attributed it to the shortness. [2] Introduction of Cr is considered as another way to eliminate the negative coupling, and enhancement of $T_C$ in R$_2$(Fe, Cr)$_{17}$ by Cr has been explained along this line. [3, 4]

Role of the boron in Nd$_2$Fe$_{14}$B has also attracted attention. Kanamori proposed a concept called cobaltization, in which Fe atoms are made Co-like by boron or another element. [5] Tatetsu et al. have recently reported that the effect from cobaltization is unfavorable to the magnetization, although they let its relation to $T_C$ remain open. [6]

In this paper, we calculate inter-site magnetic couplings for Sm$_2$Fe$_{17}$ and Nd$_2$Fe$_{14}$X (X = B, C, N, O, F) using Liechtenstein’s formula based on first principles to tackle the questions above.
Contrary to the previous results [2], the magnetic coupling between Fe atoms with the shortest bond in Sm$_2$Fe$_{17}$ is found positive in our calculation. The conclusion of Li et al. could be a misattribution of negative couplings of longer bonds. We also discuss $T_c$ for R$_2$(Fe, Cr)$_{17}$ on the basis of our previous result. [7]

As for $T_c$ of Nd$_2$Fe$_{14}$B, we consider metastable Nd$_2$Fe$_{14}$ as a reference. By separating chemical effects from the effect of volume changes, we show that there is a positive contribution to $T_c$ from the chemical effect of B. We also show a comparison with the effect of X in Nd$_2$Fe$_{14}$X (X = C, N, O, F) to see the chemical trends.

![Figure 1](image)

**Fig. 1** Inter-site magnetic couplings between Fe-Fe pairs in Sm$_2$Fe$_{17}$ as a function of the length of the bond.
Fig. 2 Curie temperature calculated within the mean-field approximation for 
$\text{Nd}_2\text{Fe}_{14}X$ (X = B, C, N, O, F) and $\text{Nd}_2\text{Fe}_{14}$ (X = Empty).

[1] M. Sagawa, JAPAN PRIZE 2012 Commemorative Lectures. 
http://www.japanprize.jp/data/prize/commemorative_lec_2012_e.pdf
[2] Z. W. Li and A. H. Morrish, Phys. Rev. B 55 (1997) 3670.
[3] Y. M. Hao et al., J. Phys.: Cond. Matt. 8 (1996) 1321.
[4] E. Girt, Z. Altounian and J. Yang, J. Appl. Phys. 81 (1997) 5118.
[5] J. Kanamori. Prog. Theor. Phys. Suppl. 101 (1990) 1.
[6] Y. Tatetsu et al., Phys. Rev. Materials. (to appear).
[7] T. Fukazawa et al., J. Phys. Soc. Jpn. 87 (2018) 044706.