first-principles calculation of exchange coupling constants and investigation of interface magnetism for various phases and their interfaces of permanent magnets

Asako TERASAWA
Department of Materials Science and Engineering, Tokyo Institute of Technology
J1-3, Nagatsuta-cho 4259, Midori-ku, Yokohama 226-8502, Japan

Improvement of Nd-Fe-B magnets without heavy rare earth elements is one of the topics of increasing importance in the applied physics and materials science. The key for high-coercivity Nd-Fe-B magnets lies in the Nd-rich grain boundary (GB) phases [1]. However, the details of structural and magnetic properties of those GB phases are not clarified yet. Some experiments revealed the relationship between the crystallinity of GB phases and the relative angles between the interfaces and the c-plane of neighboring grains [2], which indicates the complexity of the physics in grain boundary phases in Nd-Fe-B magnets. Particularly, we targeted the exchange couplings inside and between the various phases for a computational study of magnetism in Nd-based permanent magnets.

For this purpose, we developed "jx", a post-process program for OpenMX [3] to calculate exchange coupling constants between atoms based on the Liechtenstein formula [4]. The details of the implementation is explained in the published paper of [5]. During the development of jx, we found a remarkable problem about the combination of the Liechtenstein method and the linear combination of atomic orbitals (LCAO) approximation. When adopting diagonal elements of non-orthogonal (NO) Hamiltonian as effective single-site potentials, the calculated exchange coupling constants do not converge as increasing the number of basis. Figure 1(a) shows the exchange coupling constants J_{ij} as functions of atomic distance r_{ij} for different choice of basis sets. Here, the notation sxpydzfw means that the basis set is constructed from x types of s orbitals, y types of p orbitals, z types of d orbitals, and w types of f orbitals. It is possible to see in Fig. 1 that the J_{ij} profiles are similar for s2p2d1, s2p2d2, and s3p2d2 with fluctuations of about a few meV, whereas the J_{ij} profiles deviate strongly to negative values for the larger basis sets.

To solve this problem, we introduced a new scheme to orthogonalize the atomic orbitals, namely the single-site orthogonalization (SO) scheme. In the SO scheme, we derive a basis set $\{|i\rangle_i\}$ to orthogonalize only i-th element from the original non-orthogonal orbitals $\{|i\rangle\}$. The single-site orthogonlized basis set is defined by the following equations:

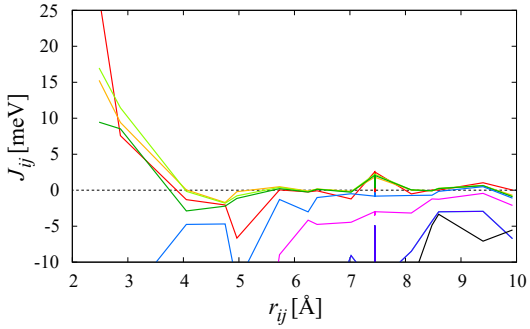
$$|i\rangle_i \equiv |i\rangle - \sum_{j\in\bar{i}} |j\rangle [\mathbf{S}^{-1}_{\bar{i},\bar{i}}\mathbf{S}_{\bar{i},i}]_{ji} \quad (1)$$

$$|j\rangle_i \equiv |j\rangle,\ j \neq i \quad (2)$$

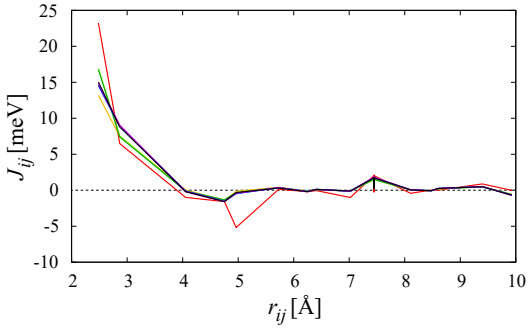
with the definitions of the neighbor set $\bar{i}$ for site i as

$$\bar{i} \equiv \{j \,|\, j \neq i,\ \langle i|j\rangle \neq 0\}. \quad (3)$$

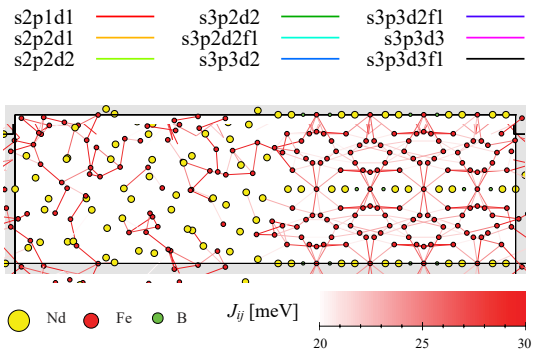
We found that the J_{ij} profiles calculated with the SO scheme exhibit a convergent behavior as increasing the number of bases, while they



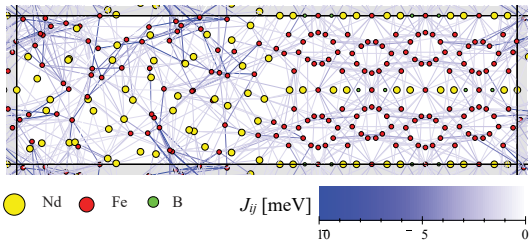
(a) J_{ij} of bcc Fe by SO scheme



(b) J_{ij} of bcc Fe by SOS scheme



(c) large positive J_{ij} in the ab interface model



(d) negative J_{ij} in the ab interface model

Figure 1: Calculated exchange coupling constants J_{ij} of various systems. (a) J_{ij} of bcc Fe as functions of atomic distance r_{ij} for different choices of basis sets when adopting diagonal elements of NO Hamiltonian as effective single-site potentials and (b) when adopting diagonal elements of SO Hamiltonian as effective single-site potentials and applying the spin population scaling to the SO results. (c) and (d) J_{ij} in the ab interface model of Nd-Fe-B permanent magnet.

decreases slightly depending on the basis size. The slight decreases in SO results are well collected by introducing spin population scaling, namely single-site orthogonalization with spin population scaling (SOS) scheme:

$$J_{ij}^{(\mathrm{SOS})} \equiv \frac{\Delta n_i}{\Delta n_j^{(\mathrm{SO})}} \frac{\Delta n_j}{\Delta n_j^{(\mathrm{SO})}} J_{ij}^{(\mathrm{SO})}. \qquad (4)$$

where $J_{ij}^{(\mathrm{SO})}$ is the exchange coupling constant derived by the SO scheme, $\Delta n_i^{(\mathrm{SO})}$ and $\Delta n_i^{(\mathrm{SO})}$ are the spin population at site i calculated using the NO basis and SO basis, respectively. Figure 1 (b) shows the exchange coupling constants as functions of atomic distances calculated with the SOS scheme. We can see a remarkable convergence as increasing the number of bases in Fig. 1(b).

We also applied the newly developed scheme to larger scale systems. Figures 1(c) and 1(d) show the calculated exchange coupling constants of the model of ab interface, where the amorphous Nd-Fe phase of a few nm are attached to the main phase in ab axis direction. It can be seen in Figs. 1(c) and (d) that the exchange coupling constants behave differently in the main phase and in the amorphous Nd-Fe phase.

We are now preparing the submission of the above results to refereed papers, and modified patch of jx including the SOS scheme for future release of OpenMX.

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