First-Principles Simulation of $Nd_2Fe_{14}B$/Nd-Rich Phases

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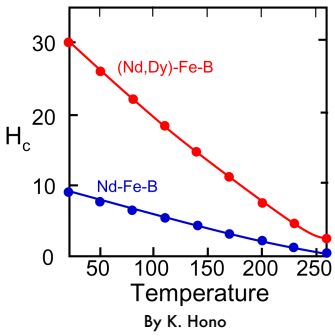
Introduction

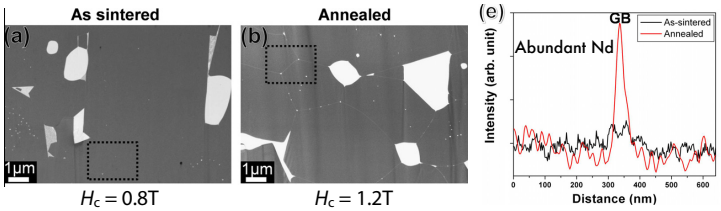
・Critical problem of Nd-Fe-B sintered magnet



The ideal coercivity ~ 7.7T
↓
Very small coercivity: H_c ~ 1.2T
↓
Dy doping → H_c~ 3 T
↓
Problems in terms of elements strategy and physics

- Elements strategy: Dy should not be used
- Physics: Why H_c is so small and what determines H_c ?

・Grain boundary is important for enhancing coercivity



H_c = 0.8T H_c = 1.2T

H. S.-Amin et al., Acta Materialia **60**, 819 (2012).

Clear Nd-rich grain boundary is made by annealing
↓
Coercivity H_c becomes higher
↓
Objective
Understanding of coercivity microscopically: to clarify the electronic structures in grain/GB interface

Simulation Methods

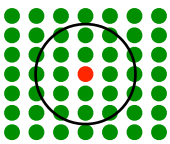
・Electronic structure calculation: density functional theory

VASP code:
- Plane wave basis
- PAW method
- GGA-PBE exchange correlation functional
- GGA+U method for 4f orbitals in Nd (U = 6eV)
- 4f orbitals of Nd in the core for structure optimization

・Large scale simulation

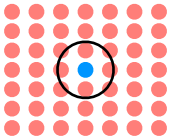
OpenMX code:
- Optimized pseudo-atomic orbital basis
- Pseudopotential method
- GGA-PBE exchange correlation functional
- GGA+U method for 4f orbitals in Nd (U = 6eV)
- Krylov-subspace order-*N* method

Original space

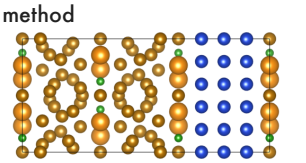


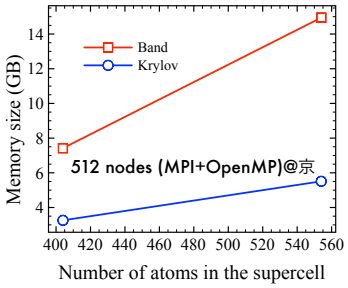
basis: $|W_0\rangle = (|i1\rangle, |i2\rangle, \cdots, |iM_i\rangle)$

Krylov subspace



basis: $U_K = \{|W_0\rangle, A|W_0\rangle, A^2|W_0\rangle, \cdots, A^q|W_0\rangle\}$
$A = (S^{(i)})^{-1}H^{(i)}$

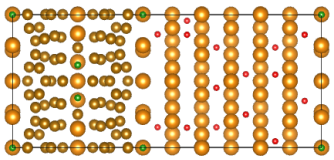




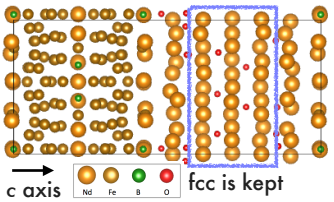
Numerical Results

・$Nd_2Fe_{14}B$/Nd_4O (VASP simulation)
- $Nd_2Fe_{14}B$: $\sqrt{2}\times\sqrt{2}$; Nd_4O: $\sqrt{5}\times\sqrt{5}$
- Lattice mismatch: 1.2%

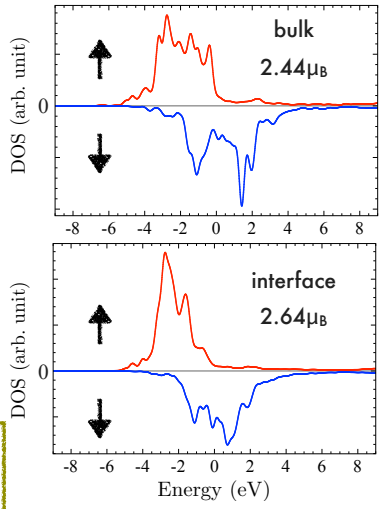
Before structure optimization



After structure optimization



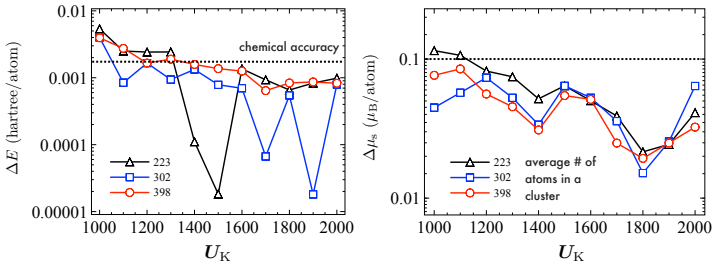
Density of states for *d*-orbital of Fe



- The electronic states in the bulk and at the interface are drastically different
- Spin moment is large at the interface

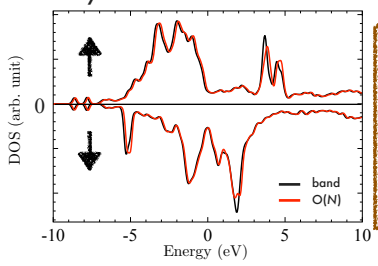
・Order-*N* simulation of $Nd_2Fe_{14}B$ (OpenMX simulation)

Confirmation of accuracy of O(*N*) calculation



- The chemical accuracy is accomplished even for small U_K.
- When the number of atoms in a cluster is small, the accuracy widely fluctuates with increasing U_K.

Density of states: band calculation vs. O(*N*) calculation



The density of states is reproduced using the information in O(*N*) simulation

Future
Application to large-scale systems: $Nd_2Fe_{14}B$/Nd-rich phase

Summary

$Nd_2Fe_{14}B$/Nd_4O
- After the structure optimization, the fcc structure of Nd_4O is kept
- The interface electronic states and spin moment of Fe are totally different from those in the bulk

Order-*N* calculation on $Nd_2Fe_{14}B$
- The Krylov-subspace method can reach the chemical accuracy by significantly smaller computational cost than conventional divided-conquer methods
- We determined appropriate parameters such as the cluster size and the Krylov dimension, for $Nd_2Fe_{14}B$/Nd-rich phase simulations

Acknowledgements

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