永久磁石における材料組織効果の第一原理電子論

Microstructure effects in permanent magnets studied by first-principles electron theory

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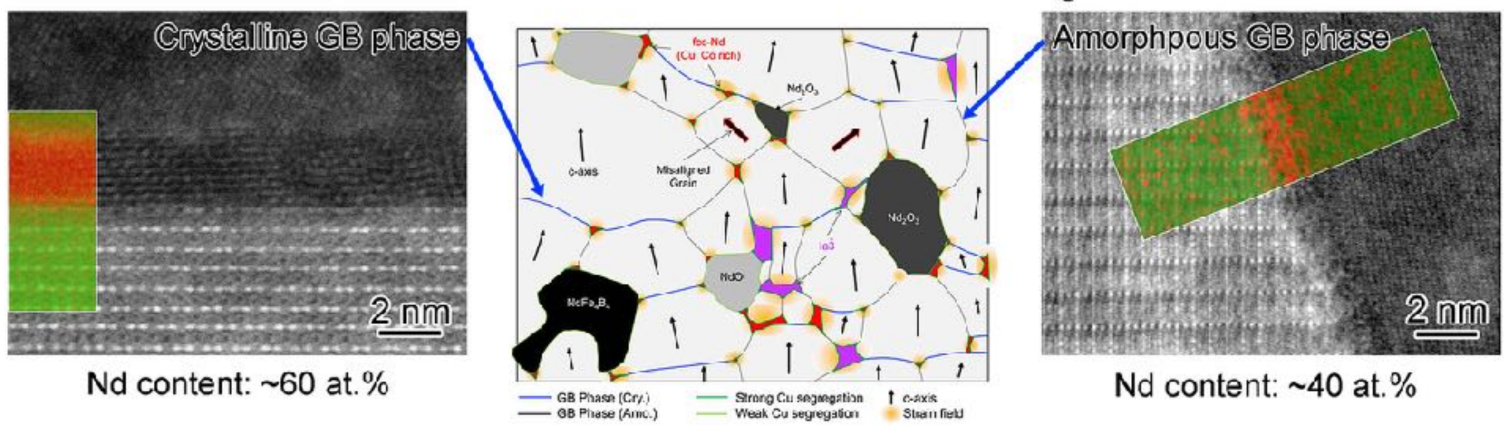
要旨

第一原理電子論により、①フォノンと磁性の相関を考慮した有限温度磁性の理論スキームを構築した。②CALPHADデータベースと第一原理計算を組み合わせた状態図の高精度化によりSmCu液相の存在を示し、1-12系主相/SmCu副相界面の局所磁気特性を明らかにした。③「京」などを用いた大規模第一原理計算により、ネオジム磁石における偏在Cuの効果や主相–副相界面におけるGa添加効果、主相における軽元素の役割を議論した。

Introduction

Microstructure of Nd-Fe-B sintered magnet



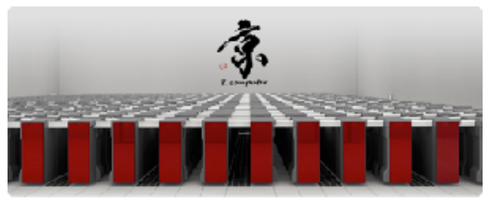
Nd content: ~60 at.%　　Nd content: ~40 at.%

T. T. Sasaki *et al.*, Acta Mater. **115**, 269 (2016).

Phase equilibria?

Atomic arrangements and influences on local magnetic properties?

Methods



First-principles calculations based on DFT-GGA-PBE

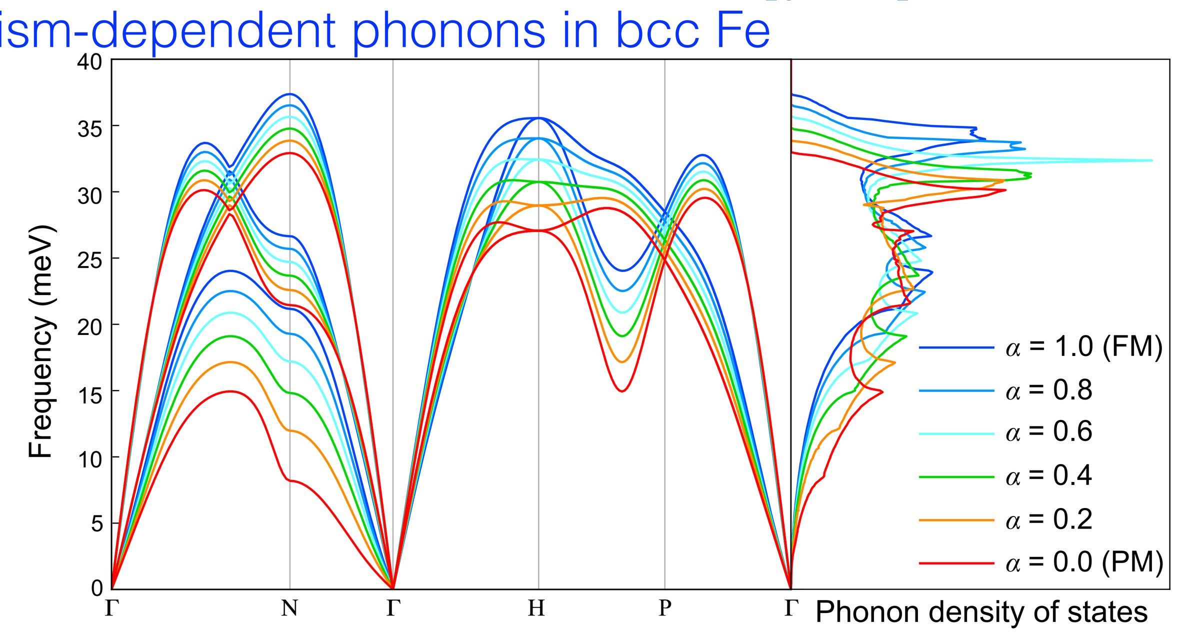
- Open-core pseudopotential for Nd
- Phonon calculations by ALAMODE
- Crystal-field analysis for magnetic anisotropy (K_1) of Nd
- Perturbation formalism for anisotropy of Fe

Torbatian, Ozaki, Tsuneyuki, & Gohda, Appl. Phys. Lett. **104**, 242403 (2014).

Finite-temperature magnetism

T. Tanaka and Y. Gohda, npj Comput. Mater. **6**, 184 (2020).

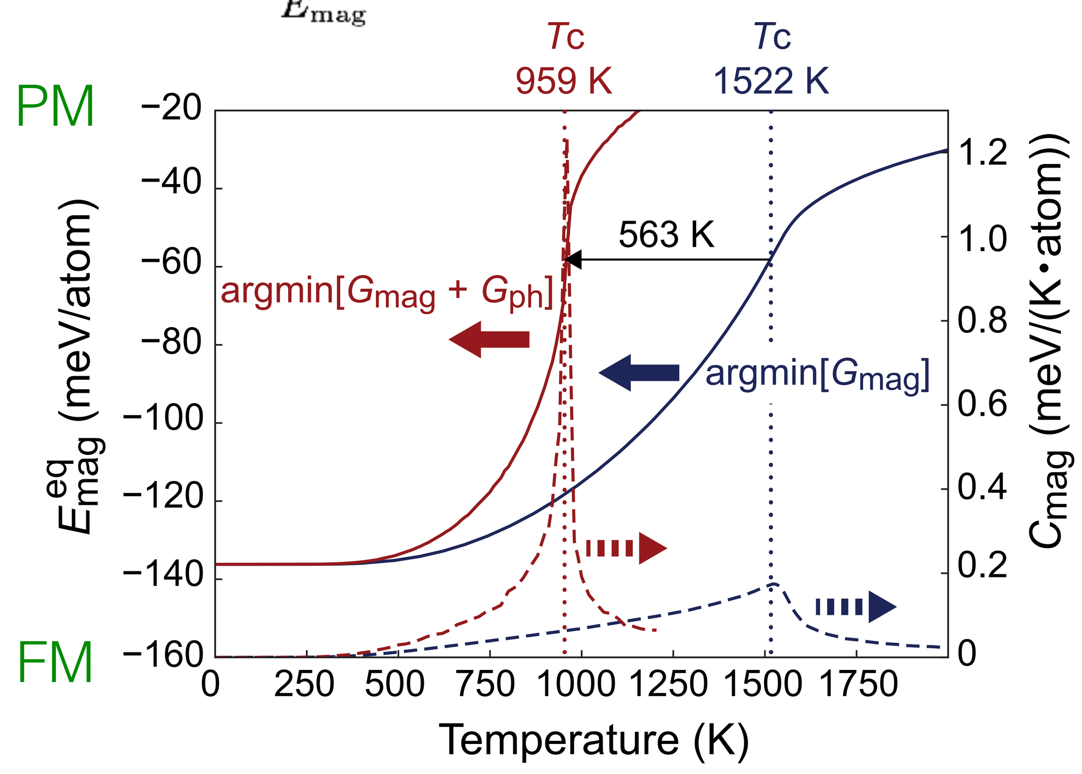
magnetism-dependent phonons in bcc Fe



$$G^{\mathrm{excess}}_{\mathrm{mag}}(T) = G_{\mathrm{Heisenberg}}(\{J_{ij}(T, E_{\mathrm{mag}}(T))\}) + \Delta G_{\mathrm{ph}}(E_{\mathrm{mag}}(T), T)$$

(approximated for fixed volume)

$$E^{\mathrm{eq}}_{\mathrm{mag}}(T) = \underset{E_{\mathrm{mag}}}{\mathrm{argmin}}\,[G_{\mathrm{mag}}(T, E_{\mathrm{mag}}) + G_{\mathrm{ph}}(T, E_{\mathrm{mag}})]$$



decrease in the Curie temperature by more than 500 K due to the feedback effect from magnetism-dependent phonons to magnetism

$SmFe_{12}$-based magnets

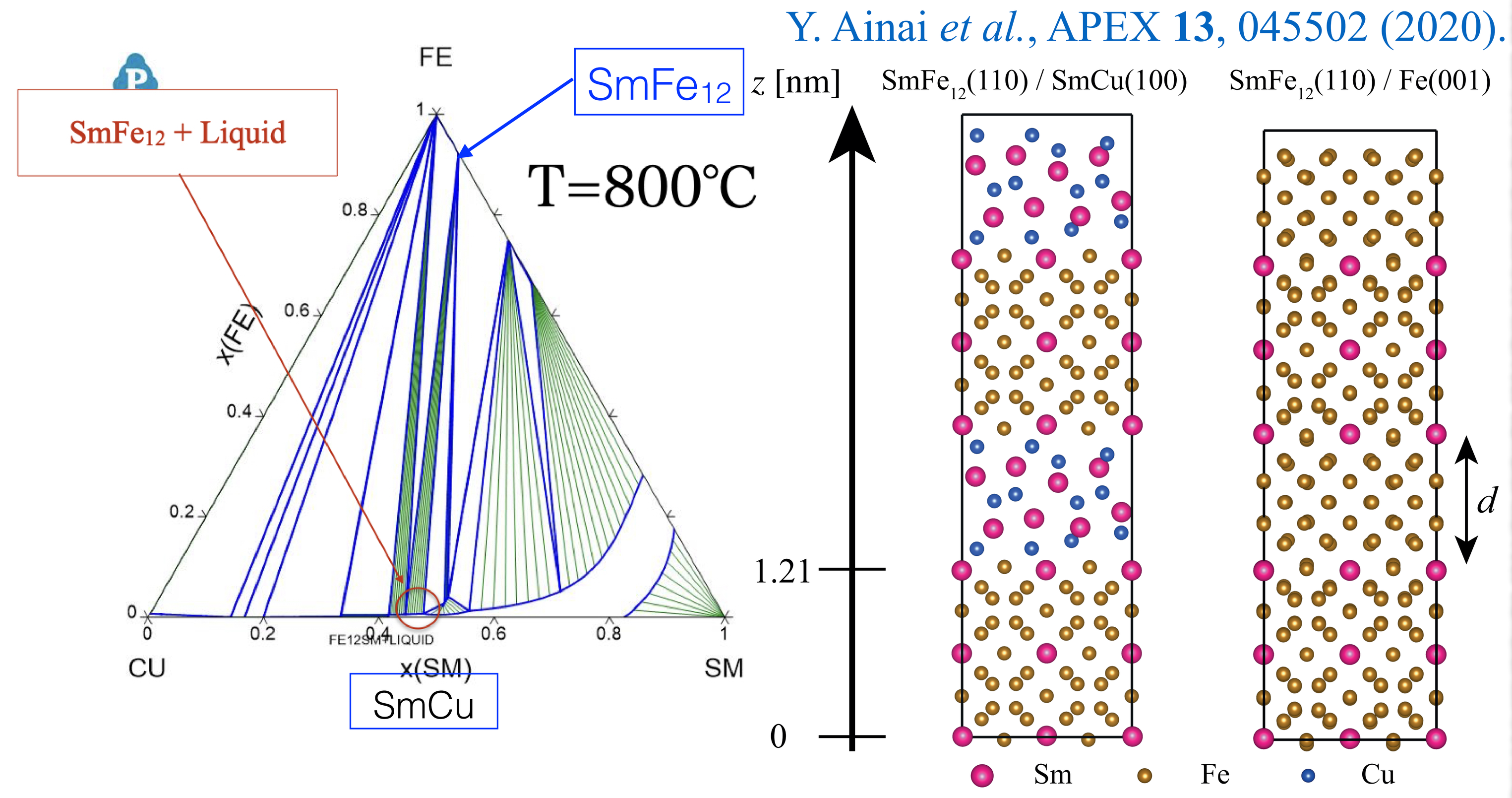
CALPHAD approach assisted by first-principles calculations

$$G^{\alpha} = \sum_{i=A}^{N} x_i G_i^{\alpha} + RT \sum_{i=A}^{N} x_i \ln x_i + G^{\alpha}_{\mathrm{excess}}$$

database　　first-principles calc.

$$G^{\alpha}_{\mathrm{excess}} = \sum_{i=A}^{N}\sum_{j>i} x_i x_j \Omega_{ij} = \sum_{i=A}^{N}\sum_{j>i} x_i x_j \left[\sum_{n=0}^{v} L^{(n)}_{i,j}(x_i - x_j)^n\right]$$

Y. Ainai *et al.*, APEX **13**, 045502 (2020).



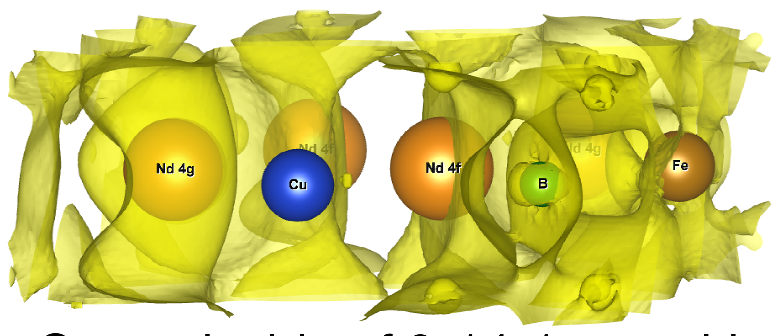
- SmCu as nonmagnetic subphase
- exchange coupling of 1-12/SmCu/1-12: 8.4% of 1-12/Fe/1-12

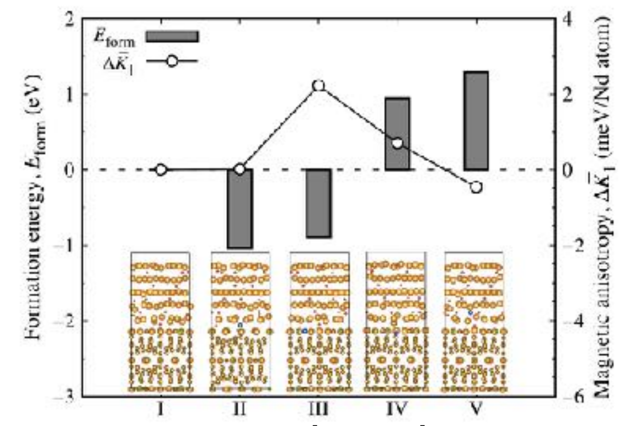
$Nd_2Fe_{14}B$-based magnets

effects of added Cu

Phys. Rev. Appl. **6**, 064029 (2016).

Y. Gohda *et al.*, Mater. Trans. **59**, 332 (2018).





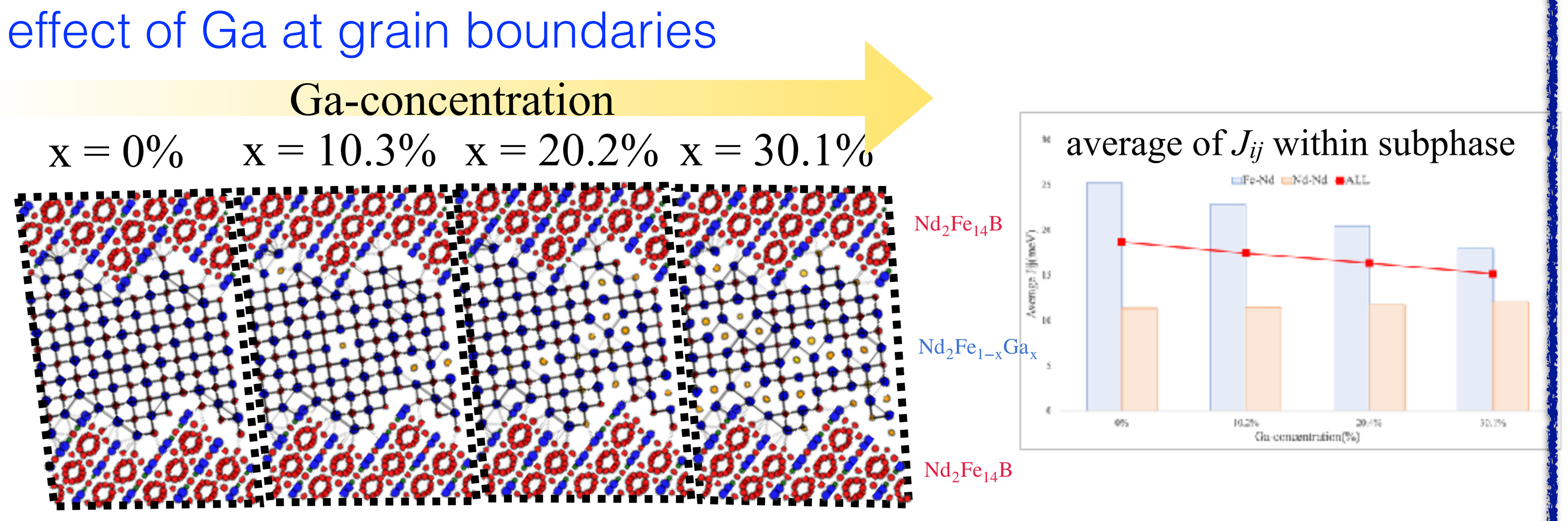
Cu not inside of 2-14-1 + positive effect on magnetic anisotropy

efficient computation of exchange couplings J_{ij}

Terasawa, Matsumoto, Ozaki, & Gohda, J. Phys. Soc. Jpn. 88, 114706 (2019).

$$J_{i\mathbf{0},j\mathbf{R}} = \frac{1}{2}\sum_{p=1}^{N_P}\tilde{R}_p \sum_{\mu,\nu\in i}\sum_{\mu',\nu'\in j} \mathrm{Re}\left\{[\hat{P}_i]_{\nu\mu} G^{+}_{i\mu,j\nu'}(\downarrow,\tilde{z}_p,\mathbf{R})[\hat{P}_j]_{\nu'\mu'} G^{+}_{j\mu',i\nu}(\uparrow,\tilde{z}_p,-\mathbf{R})\right\}.$$

effect of Ga at grain boundaries



- decrease in exchange couplings by added Ga
- local magnetic properties as inputs in spin-lattice model

role of B in 2-14-1 phase

stabilization of crystal structure
not enhancement of magnetism

Phys. Rev. Mater. 2, 074410 (2018).