Designing permanent magnets from first-principles

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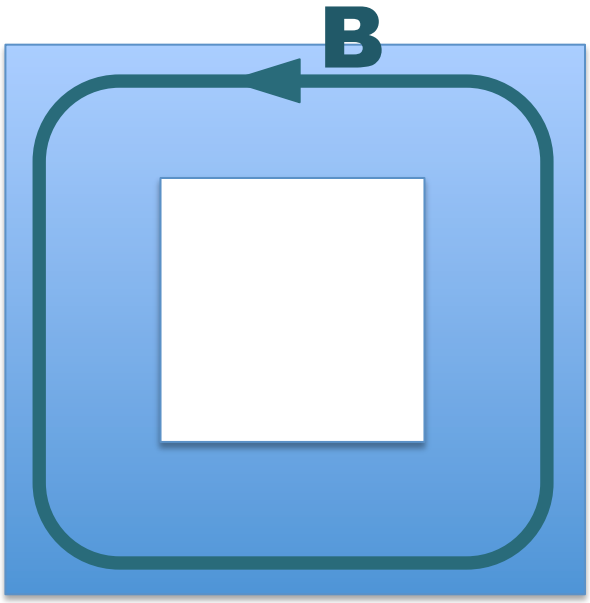
Permanent magnets (PM)

- Magnetization
- Coercivity
- Temperature characteristics

How to use magnetic field?

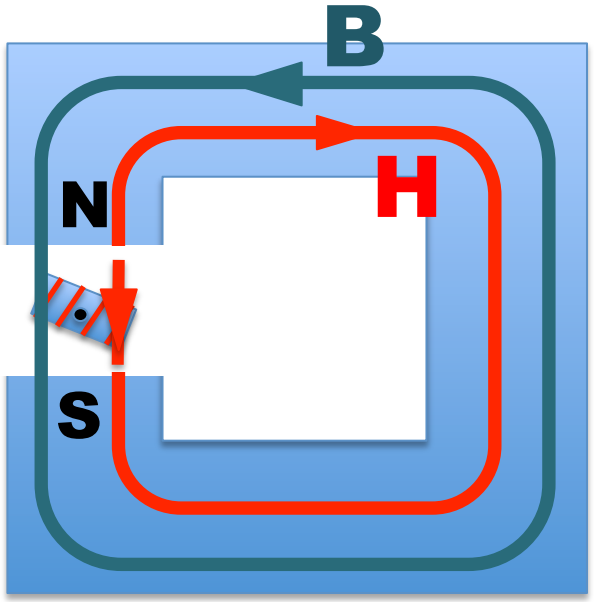
- Using magnetic field produced by magnets

Closed magnetic circuit



Magnetic field cannot be used

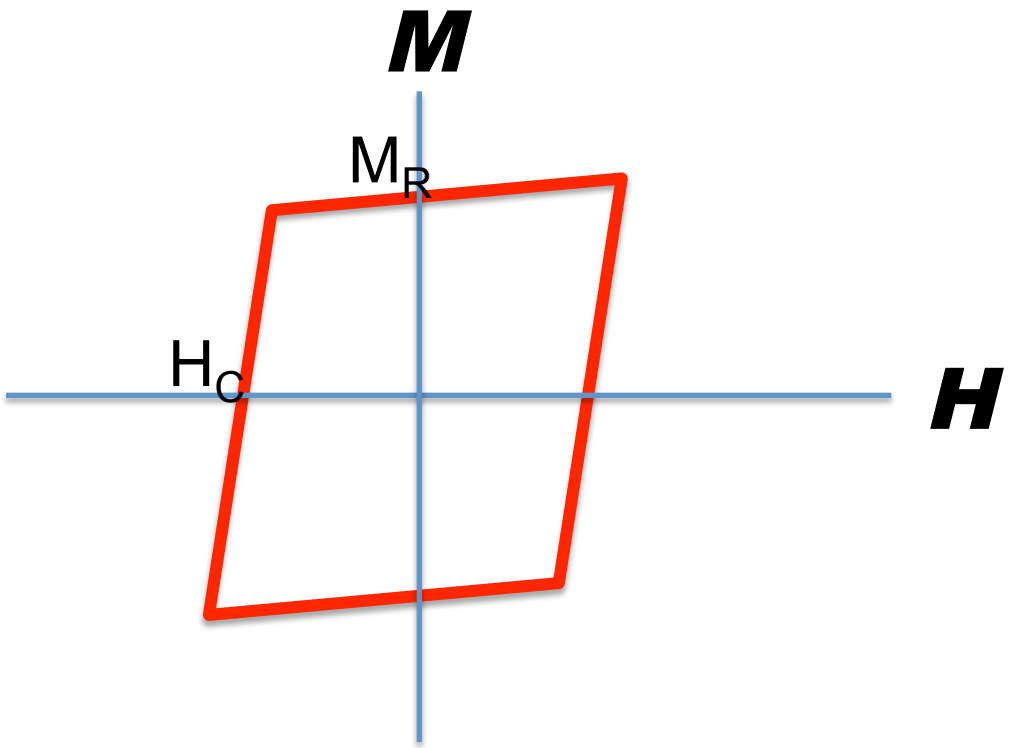
Actual situation



Magnet is under magnetic field

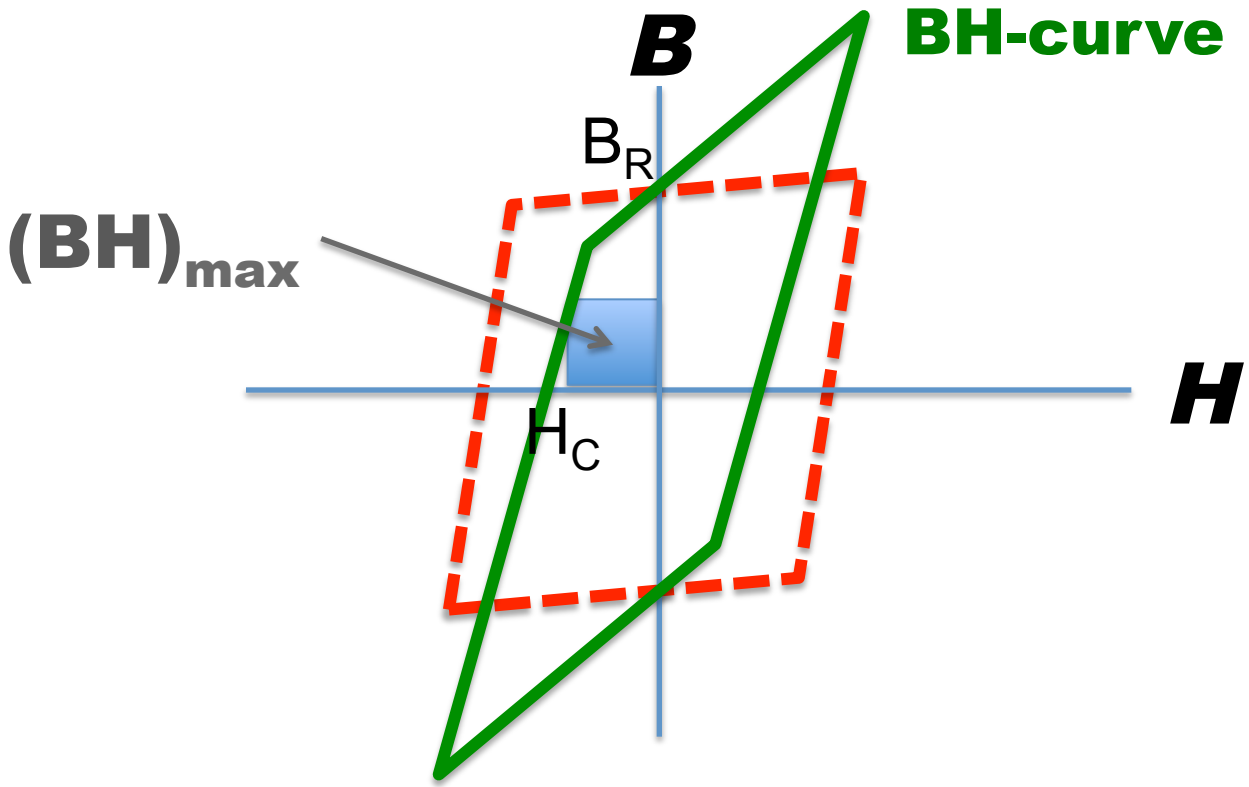
PM = ferro + coercivity

- PM is used at the second quadrant
- Without coercivity, PM cannot be used.



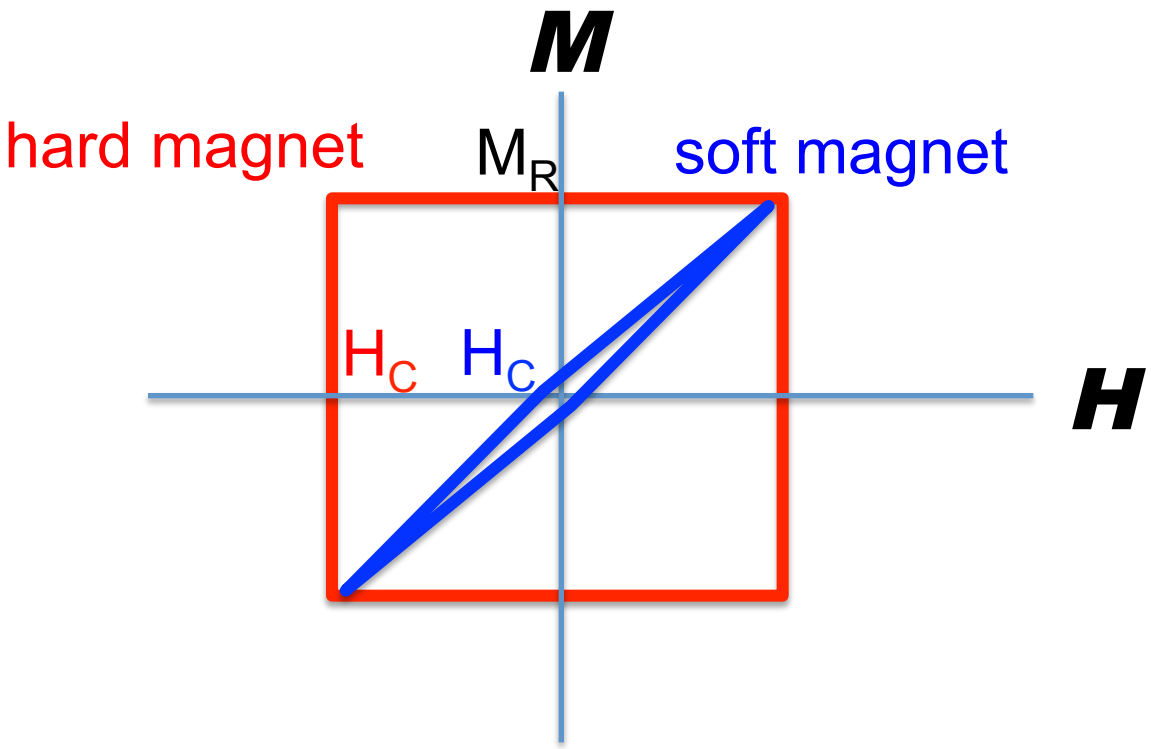
Maximum energy product

- $B=M+\mu_0 H$



Ideal PM

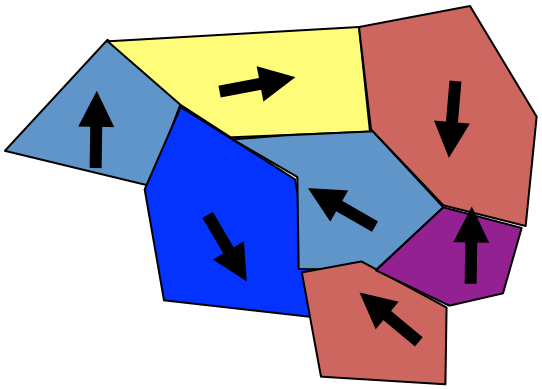
- Hard PM and soft PM

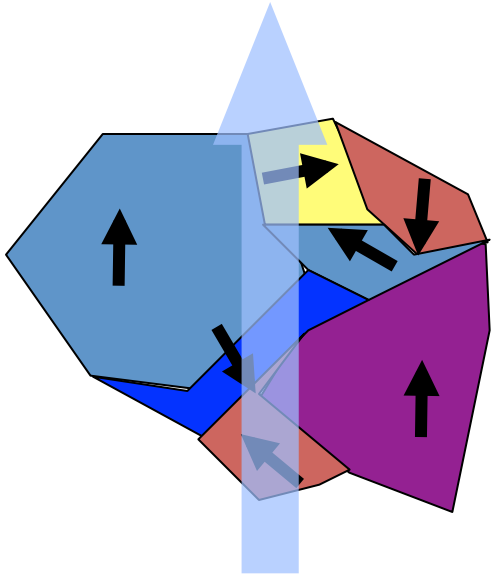


H_c is much smaller than magnetic anisotropy.

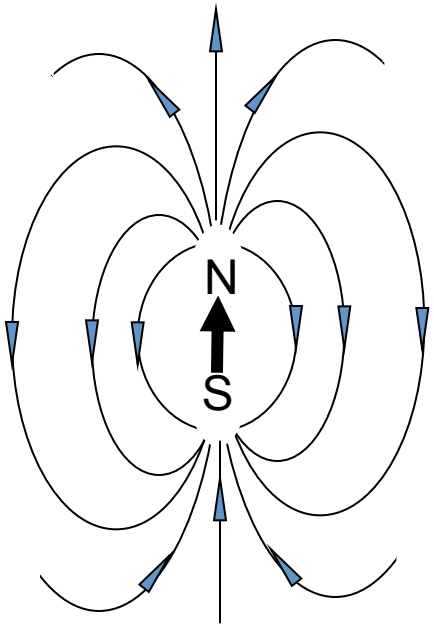
Magnetic domain

- Domain structures
- Initially no net magnetization
- Under field, preferable domain expands





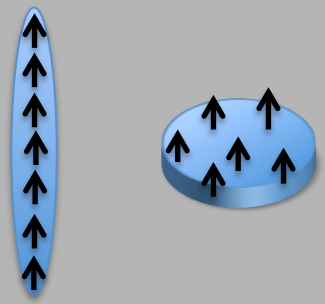
Field



Magnetic field

Hard PM

- Strong magnetic anisotropy
  - Crystalline magnetic anisotropy
  - Shape magnetic anisotropy
- Magnetic domain structure
  - Pinning of domain wall
  - Single domain structure



- Single-domain structure →fine particles
- Acicular crystal →spinodal decomposition
- Pinning →defects
- Spin-orbit coupling →rare-earth
- Orbital moment →orbital degeneracy

Designing permanent magnets from first-principles

- How?
- Possible?
- Present status?

First-principles electronic structure calculation?

- Based on the density functional theory (DFT)
- Ab-initio, yet approximations needed for E_{xc}
  - Local approximation (LDA, GGA, …)
  - Local + something (SIC, hybrid, …)
  - A bit better approaches (EXX-OEP, EXX+RPA, GW, …)
- Mainly for crystals, i.e. periodic systems
  - APW, KKR, their linearized versions, pseudo-potential, …
- Various kinds of extension
  - Impurity, disorder, clusters, multilayers, surfaces, interfaces,..
  - Structural optimization, ab-initio molecular dynamics,…

What can be discussed?

- Properties at ground state
  - Electron density n, spin density m
  - Quantities that are functional of n and m
    - total energy, magnetization, electrostatic potential, …
  - First order perturbation to the total energy
    - static susceptibility, hyperfine fields, isomer shifts
  - Ground state under constraint
    - $E[m]$, …
  - Second order perturbation such as susceptibility, conductivity, … – not completely justifiable, marginal

Slightly beyond DFT –yet acceptable

- Local susceptibility χ_{ij}、exchange constants J_{ij}

$$J_{ij} = \frac{1}{\chi_{ii}} \chi_{ij} \frac{1}{\chi_{jj}}$$

- Atomic pair interactions
- Conductivity σ、optical conductivity $\sigma_{\mu\nu}(\omega)$
- Temperature effects（through Fermi distribution function）
- Combination with many-body perturbation theories

Designing permanent magnets

- First step: simulation of magnetic systems
- Second step: simulation of permanent magnets
- Third step: design permanent magnets

First step
– simulation of magnetic systems

- Magnetization
- Curie temperature
- Crystal magnetic anisotropy

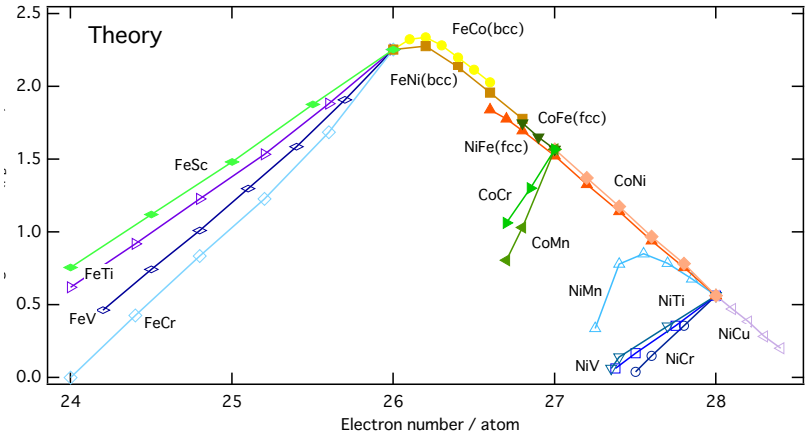
Magnetization

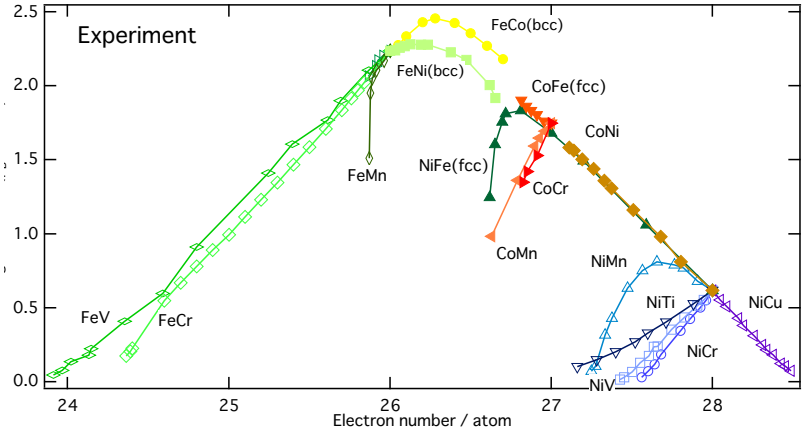
- Accurate enough for materials used for permanent magnets

Slater-Pauling curves (1991)

Magnetization of 3d disordered alloys

KKR-CPA-LDA





Well described within DFT-LDA

Curie temperature

- Method applicable for which local magnetic moment well developed

Calculation of J_{ij}

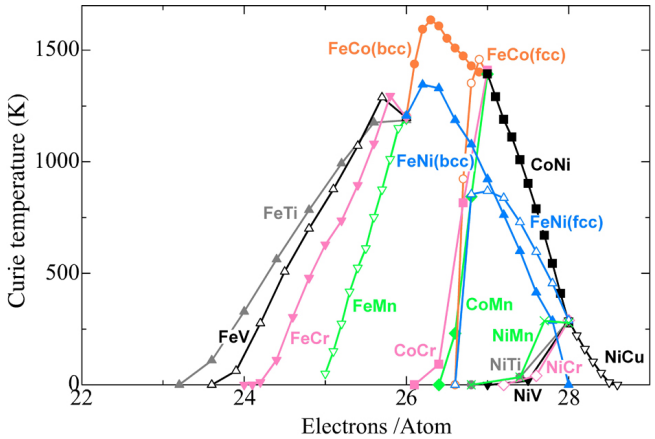
⬇

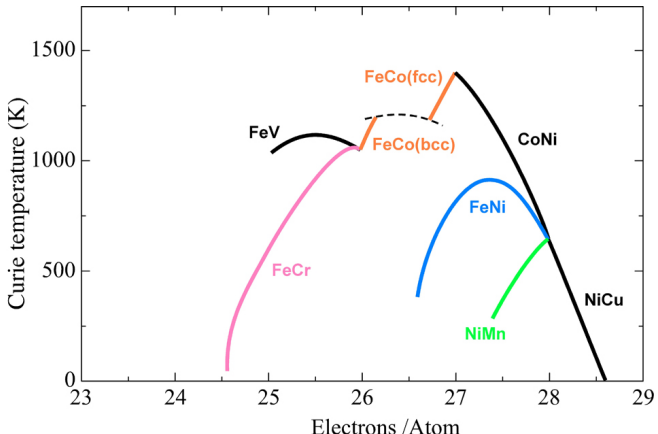
Map to Heisenberg model

⬇

Apply statistical method

Curie temperature Slater-Pauling curves





C. Takahashi et al
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Crystalline magnetic anisotropy of transition metals

- Framework of DFT (More or less)
  - spin-orbit coupling
- Cubic phase –numerically difficult

Total energy	~1000Ry
Cohesive energy	~0.3Ry
Magnetic energy	~0.02Ry
Crystal structure energy	~0.02Ry
Magnetic structure energy	~0.01Ry

Crystalline magnetic anisotropy energy
0.0000001 ~ 0.00001Ry

Numerical precision of theory
10^{-6}Ry

MAE of rare-earths

- LDA not works well
  - self-interaction ≅ exchange
  - Introducing “orbitals” needed
  - SIC(self-interaction correction) a better choice
  - LDA+U (i.e. Hartree-Fock approximation) not preferable
- Spin-orbit coupling
- Hybridization of f-orbital yet important
- Less numerical difficulty

MAE and orbital degeneracy

- Unquenched orbital moment
- Hardly occurs in metals except for rare earths
  - Narrow bands comparable to spin-orbit splitting
  - $Li_2(Li_{0.7}Fe_{0.3})N$

$Li_2(Li_{0.7}Fe_{0.3})N$

MAE field 2200kOe, coercivity field 120kOe

Aims at this stage

- Reproduce basic properties of magnetic materials quantitatively–quantum simulation
- Design new magnetic materials–CMD
  - Larger magnetization
  - Higher Curie temperature
  - Strong magnetic anisotropy
- Design preferable materials for permanent magnets
  - No rare-earth metals
  - No environmental problem
  - Low price

Next step –design permanent magnets

- Coercivity mechanism
  - Phenomenological description ⇒ microscopic theory
  - How to realize a magnetic single-domain
  - Spinodal decomposition
  - Possible sintering process
  - Grains, grain surfaces, and related properties
  - Grain boundary
  - Pinning mechanism of domain walls
- High temperature properties
  - Predictive power without relying on specific models

Some examples

- Method
  - Local density approximation of DFT
  - KKR Green's function method
- Calculation of J_{ij}
- MAE (magnetic anisotropy energy) Slater-Pauling curves
- MAE of SmFeN

Summary

- Towards designing permanent magnets
  - 1st step description of magnetic materials
  - 2nd step description of permanent magnets
  - 3rd step designing permanent magnets
- Overview of the first-principles approach
  - J_{ij}
  - T_c
  - K_1