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FIRST-PRINCIPLES CALCULATIONS OF THE 3D MAGNETO-CRYSTALLINE ANISOTROPY ENERGY OF TRANSITION METALS

IZRA^UN ENERGIJE MAGNETNO-KRISTALNE ANIZOTROPIJE ELEMENTOV PREHODA

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Mag neto-crystalline ani so tropy en ergy (MAE) of the tran si tion-metal el e ments is due to the spin-orbit (SO) cou pling of the 3d electrons. It has been studied within the framework of the local spin density approximation by the full-potential lin ear-augmented-plane-waves (FPLAPW) method. The SO cou pling is treated non self-consistently in the last it er a tion as a perturbation. The 3d MAE is determined applying the force theorem as the difference between the sum of the occupied sin gle-state en er gies for the two mag ne ti za tion di rec tions. The con ver gency of the cal c u lated MAE with re spect to the num ber of **k**-vec tors in the Brillouin zone (BZ) is rather poor when the per turbed oc cu pied states are de te r mined just ac cord ing to their en er gies and an ex tremely large BZ (more than 10000 **k**-vec tors) is re quired to ob tain sta ble re sults. There fore, the state track ing scheme has been fol lowed where the per turbed oc cu pied states are de fined ac cord ing to their wave func tions as re lated to the un per turbed oc cu pied states. The con ver gency of the cal cu lated MAE is im proved and less than 10000 **k**-points in a BZ are found to be suf fi cient for the sta ble cal cu la tion.

Key words: mag neto-crystalline ani so tropy, tran si tion-metal el e ments, den sity func tional t heory

Energija magnetno- kristalne anizotropije (MAE) elementov prehoda je posledica sklopitve spin-tir (SO) elektronov podlupine 3d. Izra~unana je bila v okviru pribli`ka lokalne spinske gostote z metodo Full-potential Linear-augmented-plane-waves (FLAPW). Sklopitev SO je obravnavana ne-samovsklajeno v zadnji iteraciji kot motnja. MAE elektronov 3d je dolo~ena s pomo~jo force the o rem-a, kot razlika vsot enodel~nih energij zasedenih stanj za dve orientaciji magnetizacije. Konvergenca izra~unane MAE je, glede na {tevilo to~k iz prve Brillouinove cone (BZ), precej slaba, ~e so zasede na motena stanja dolo~ena samo na osnovi njihovih energij (za stabilne rezultate je potrebnih ve~ desettiso~ vektorjev). Zato je bila uporabljena shema state track ing, kjer so motena zasedena stanja dolo~ena glede na projekcijo njihovih valovnih funkcij v nemotenem prostoru. Konveregenca je mo~no izbolj{ana in za stabilne izra~une zadostuje manj kot desettiso~ vektorjev v BZ.

Klju~ne besede: magnetno- kristalna anizotropija, elementi prehoda, teorija gostotnih funkcionalov

1 INTRODUCTION

The magneto-crystalline anisotropy energy (MAE) is one of the most interesting intrinsic properties of magnetic materials. It is important from the technological aspect as a quantity responsible for the high coercivity in permanent magnets, magneto-elasticity and magneto-recording properties. The calculation of MAE from electronic structure within the framework of the density functional theory[1] still represents a difficult task. The MAE of the transition-metal materials originates from the spin-orbit coupling (SOC) of the 3d electrons. The difficulties associated with the calculation of the 3d MAE arise from the fact that its magnitude is very small (for bulk bcc Fe ~ μeV/atom) in comparison with the total energies per unit cell of the crystal (for bulk bcc Fe ~ 10^4 eV).

The magneto-crystalline anisotropy energy E_{MAE} is defined as the difference between the total energies for the two magnetization directions defined by the azimuthal θ and polar Φ angles:

$$E_{MAE} = E(\theta', \Phi') - E(\theta'', \Phi''). \quad (1)$$

The SOC for the 3d metal system is very weak (~ 40 meV) when compared to the bandwidth (~4eV) and therefore it can be treated as a perturbation. First, the self consistent calculation without SOC is performed. The SOC for the two directions of the magnetization is then applied non self-consistently in a perturbative way. According to the force theorem[2], E_{MAE} can be approximated as the difference between the sums of the eigenvalues of the perturbed occupied states over the Brillouin zone (BZ):

$$E_{MAE} \approx \sum_{(k,i)\in\{occ'\}} \varepsilon_i''(k) - \sum_{(k,i)\in\{occ''\}} \varepsilon_i''(k) ,$$

where i is the band index and {occ'} and {occ"} denote the sets of the perturbed occupied states for the two magnetization directions. The force theorem is valid in the limit where the charge densities obtained after applying the SOC do not differ significantly from the charge density obtained self-consistently. The notorious prob lem with such an approach is the bad convergency of the E_{MAE} with respect to the number of k-vectors (N_k) in the BZ. Therefore a very large N_k is required in order to obtain stable and reasonable results in agreement with experimental data. For example: Daalderop et al. [2] used ~ 100^3 k-vectors in the full BZ for the calculation of MAE in bulk transition metals. Furthermore, their results do not quite reproduce the experimental values.

Wang et al.[3] showed that the main reason for the numerical instability was the Fermi filling scheme where the sets of the occupied perturbed states {occ'} and {occ"} are determined just according to their eigenvalues $\varepsilon'_i(k)$, $\varepsilon''_i(k)$. The resulting differences in the charge densities are large enough to violate the basic requirement of the force theorem. They proposed the state tracking approach in which the occupied perturbed states {occ'} and {occ"} are determined according to their projections back to the set of the unperturbed states which is the result of the self-consistent calculation without SOC. This ensures minimum changes of the charge densities which is the basic condition for the application of the force theorem. State tracking was proved in practice to be a stable and precise tool for the calculation of the MAE of thin films[3] and strained bulk transition metals[4].

In this paper we have calculated the MAE of bulk non-strained Fe, Ni and Co in order to test the state tracking approach, applied to a system with extremely high symmetry and therefore very small MAE.

2 COMPUTATIONAL DETAILS

The calculations have been carried out for body-centered-cubic (bcc) Fe, face-center-cubic (fcc) Ni and hexagonal-closed-packed (hcp) Co. We have used the WIEN97[5] code, which adopts the full-potential linearized augmented-plane-wave (FLAPW) method[6]. For the exchange-correlation potential the form deduced by Perdew and Wang[7] was used. The self-consistent calculation was performed in the scalar-relativistic approximation[8] for the valence electrons, whereas the core electrons were treated fully relativistically. The criterion for the self-consistency was the difference in the charge densities after the last two iterations being less than 10^{-5} e/(a.u.)^3. The contribution of the SOC was determined using the second variational method[9,10] where the perturbed Hamiltonian is diagonalized in the base of the unperturbed eigenstates rather than in the base of the plane waves. The MAE was calculated by applying the force theorem using (i) the Fermi filling (ii) the state tracking approach to determine the sets of the perturbed occupied states {occ'} and {occ"}.

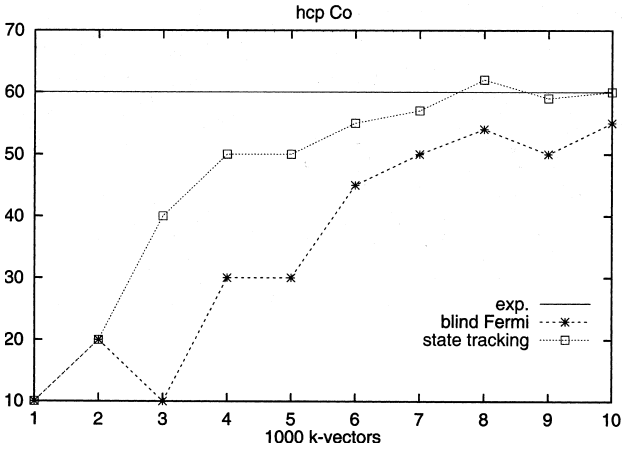

Figure 2: The cal cu lated MAE for fcc Ni with re spect to the num ber of k-vectors in units of μeV/atom. The hor i zon tal line rep re sents the experimental data

Slika 2: Izra~unana MAE v enotah μeV/atom za fcc Ni glede na {tevilo vektorjev v BZ. Vodoravna ~rta predstavlja eksperimentalno vrednost

3 RESULTS

The experimental value of the MAE for bcc Fe, defined as the energy difference between the [001] and [111] crystallographic directions, is -1.4μeV/atom[11]. **Figure 1** shows the convergency of the calculated results

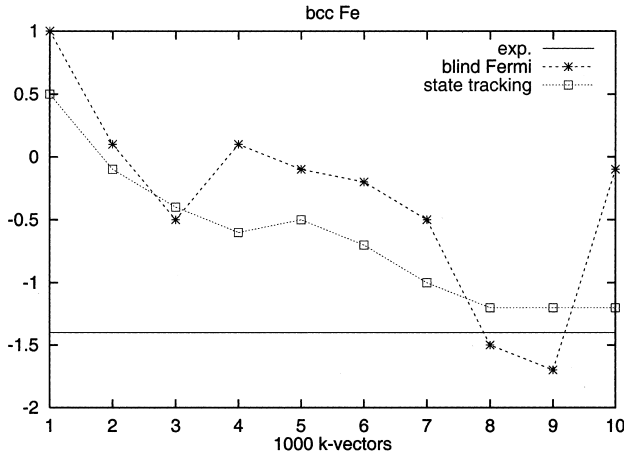

Figure 1: The cal cu lated MAE for bcc Fe with re spect to the num ber of k-vectors in units of μeV/atom. The hor i zon tal line rep re sents the experimental data

Slika 1: Izra~unana MAE v enotah μeV/atom za bcc Fe glede na {tevilo vektorjev v BZ. Vodoravna ~rta predstavlja eksperimentalno vrednost

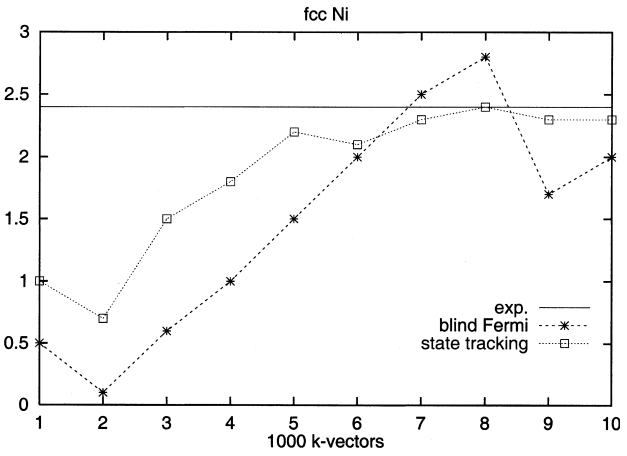

Figure 3: The cal cu lated MAE for hcp Co with re spect to the num ber of k-vectors in units of μeV/atom. The hor i zon tal line rep re sents the experimental data

Slika 3: Izra~unana MAE v enotah μeV/atom za hcp Co glede na {tevilo vektorjev v BZ. Vodoravna ~rta predstavlja eksperimentalno vrednost

with respect to the number of k-vectors in the BZ. It is obvious that the convergency of the results is very weak when the Fermi filling is applied and much more than 10000 k-vectors would be required to obtain reasonable results. In the case of the state tracking results ~ 800 k-vectors in the BZ are enough for stable results although the calculated MAE does not exactly match the experimental value. The Fermi filling does not give reasonable results in the case of fcc Ni, where the experimental value of MAE is 2.4 μeV/atom[11]: **Figure 2**. The state tracking approach provides good convergency and the calculated value is almost equal to the experimental data. The MAE of hcp Co, defined as the energy difference for the magnetization parallel and perpendicular to the c crystal axis is sufficiently large (experimental value -65μeV/atom[12] that even the results obtained according to the Fermi filling scheme almost converge with less than 10000 k-vectors although the discrepancy with the experiment is relatively large as is shown in **Figure 3**. As is expected from the results for the cubic materials, the state tracking approach almost exactly reproduces the experimental data for hexagonal Co where the MAE is one order of magnitude higher.

4 CONCLUSION

First-principles calculations on the cubic Fe and Ni and hexagonal Co systems using the FLAPW method were performed. The Fermi filling and the state tracking approach schemes were used to determine the set of the perturbed occupied states after applying the SOC. We did not obtain reliable results for the cubic system using a reasonable number of k-vectors whereas the state tracking approach gave stable results in agreement with experimental data for $N_k \approx 10000$. The symmetry of hexagonal systems is less than the symmetry of a cubic system and therefore the MAE is one order of magnitude larger. Even the Fermi filling applied to hcp Co gives relatively stable results, although the values obtained using the state tracking approach are still more reliable.

The state tracking approach has proved to be the correct scheme for the calculation of the 3d magneto-crystalline anisotropy energy of bulk transition metals.

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