



HAL
open science

MAGNETIC ANISOTROPY IN Fe, Co AND Ni

G. Daalderop, P. Kelly, M. Schuurmans, H. Jansen

► **To cite this version:**

G. Daalderop, P. Kelly, M. Schuurmans, H. Jansen. MAGNETIC ANISOTROPY IN Fe, Co AND Ni. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-93-C8-94. 10.1051/jphyscol:1988833 . jpa-00228505

HAL Id: jpa-00228505

<https://hal.science/jpa-00228505>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

MAGNETIC ANISOTROPY IN Fe, Co AND Ni

G. H. O. Daalderop, P. J. Kelly, M. F. H. Schuurmans and H. J. F. Jansen¹

Philips Research Laboratories, P.O. Box 80.000, 5600 JA Eindhoven, The Netherlands

Abstract. – Magnetic anisotropy energies are calculated *ab initio* for bcc Fe, fcc Ni and hcp Co within the local spin density approximation using the LMTO method. Although the calculated energies are in all cases of the correct order of magnitude, only in cobalt is the easy axis found to be in agreement with experiment.

Our present understanding of magneto-crystalline anisotropy in itinerant magnets is limited to a few well-studied cases such as Ni whose electronic structure is known in detail [1]. There is little available to guide experimentalists in their search for new materials with enhanced magnetic anisotropy. In particular we are interested in the magnetic anisotropy of magnetic superlattices and the mechanisms responsible for the switch of the easy axis from in-plane to out-of-plane such as occurs in the Pd/Co system when the thickness of the Co layer is less than 7.2 Å [2]. Since the electronic structures of such superlattices are not known experimentally our aim is to develop a simple and efficient *ab initio* means of calculating the magnetic anisotropy for itinerant electron systems. We do this with the Local Spin Density Approximation (LSDA) to the Density Functional Formalism, using the linear muffin tin orbital (LMTO) method within the atomic sphere approximation (ASA) [3]. In this contribution we present our first results for the elemental metals Fe, Co and Ni.

The magnetic anisotropy energy, E_A , is extremely small, of the order of $\mu\text{eV}/\text{atom}$ for Fe and Ni and approximately an order of magnitude larger for Co. To calculate such small energy differences we use the so-called force theorem [4]. This takes advantage of the variational expression for the total energy to express the change in total energy resulting from a small perturbation as the change in the sum of the single particle eigenvalues. Taking the spin-orbit coupling as the small perturbation (small with respect to the d band-width and exchange-splitting) we first perform a self-consistent semi-relativistic spin-polarized band structure calculation. Choosing the spin quantization axis along different crystallographic directions we then calculate the change in energy when the spin-orbit coupling is included. The difference between two such energy differences, $\Delta \sum_{i=\text{occ}} \delta \epsilon_i$, is the anisotropy energy E_A . All other calculations we are aware of similarly restrict their attention to the change in the sum of the single particle eigenvalues [1]. This sum is performed using the linear tetrahedron method, with the correct weighting of the tetrahedra to ensure fast convergence for filled bands [5].

Self-consistent calculations have been performed for Ni, Co and Fe at their experimental volumes. The anisotropy energies indicated in figure 1 by an arrow correspond, respectively, to $n = 10, 9$ and 8 valence electrons. For small ranges of electron numbers about these values, a rigid band model has been used to obtain $E_A(n)$. The feasibility of converging the sum to μeV precision is demonstrated in figure 1a by calculating $E_A(n)$ for Ni for divisions of the Brillouin zone into increasingly small tetrahedra. While the form of the function $E_A(n)$ may be obtained with a moderately fine division, a precise value of the anisotropy energy is only obtained in the limit that the tetrahe-

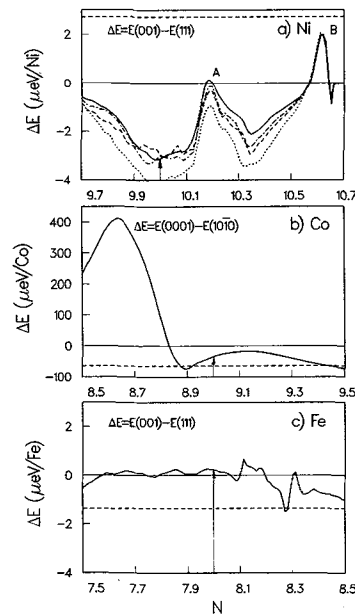


Fig. 1. – Variation of magnetic anisotropy energy with number of valence electrons (see text). The calculated magnetic anisotropy energies ($E_{001} - E_{111}$ in Figs. 1a and 1c, and $E_{0001} - E_{1010}$ in Fig. 1b) are denoted with an arrow, the experimental values are shown by the straight dashed lines. The dotted, dashed, chain-dotted and solid curves in figure 1a indicate calculations with 65, 75, 85 and 95 divisions of the reciprocal lattice vectors.

¹Dept. of Physics, Oregon State University, Corvallis, Oregon, U.S.A.

dron volume goes to zero and this value we estimate by extrapolation. For a convergence of about $0.3 \mu\text{eV}$ for Ni, we have found it necessary to use $\sim 5 \times 10^6$ tetrahedra in the full Brillouin zone. In this case the full Hamiltonian was diagonalized at a total of 120,000 irreducible k-points.

The first point to note about figure 1 is that although the calculated anisotropy values are in all three cases of the correct order of magnitude, the easy axis is only calculated correctly for Co. The experimental anisotropy energies for Ni, Co and Fe are, respectively, 2.7, -65 and $-1.4 \mu\text{eV/atom}$ [6]. We calculate them to be -3.1 ± 0.3 , -31 ± 1 , and $0.2 \pm 0.3 \mu\text{eV/atom}$. The convergence error is less than the discrepancy between the calculated and experimental values. Secondly, $E_A(n)$ is a rapidly varying function of the number of electrons n . Alloying can therefore have a large effect (if the Fermi surface is not thereby destroyed). We have verified that this rapid variation of $E_A(n)$ results from contributions associated with the lifting of degeneracies at the Fermi level by spin-orbit coupling [1]. As a consequence, the anisotropy energy does not have a simple dependence on the size of the spin-orbit coupling constant ξ . Thus for crystals with cubic or uniaxial symmetry, E_A is not necessarily proportional to ξ^4 or, respectively, to ξ^2 , which results from neglecting the degeneracies at the Fermi level [7].

The dependence of E_A on n for Ni (Fig. 1a), and in particular the peaks labelled A and B, can be understood from the electronic band structure along the symmetry lines $\Gamma-X$ and $\Gamma-L$. The mutual repulsion of bands 8 and 9 at the three X-points gives rise to peak A while peak B is due to contributions from bands 9 and 10 near these X-points. The X_2 hole-pocket, associated with empty states in band 8, is found in other LSDA calculations but not confirmed by experiment [8]. Filling this band completely leads to the displacement of peak A to the left in figure 1a thus making the calculated $E_A(n)$ less negative around $n = 10$. There is a gradual decrease of $E_A(n)$ by about $3 \mu\text{eV}$ between 9.6 and 10.0 valence electrons. This results from a cancellation of contributions from bands 8, 10, 11 and 12 which are, respectively, -1, -14, 6 and $6 \mu\text{eV}$. For a 3% volume decrease (corresponding to about 50 kBar of hydrostatic pressure) E_A increases by about $0.4 \mu\text{eV}$ around $n = 10$.

For cobalt with an ideal c/a ratio, $E_A(n)$ is shown in figure 1b. In going from 8.5 to 9.0 valence electrons there is a transition from the easy axis lying in the basal plane to it being perpendicular to the basal plane which originates from the electronic structure of the AHL-plane. The very large change in $E_A(n)$ of $400 \mu\text{eV/atom}$ is again a consequence of an "accidental" position of the Fermi-level rather than the result of a general trend. A decomposition of $E_A(n=9)$ yields a ratio of $K_1 : K_2$ of 4:3

compared to the measured ratio of 8:1 [6]. A large anisotropy in the magnetic moment per atom is computed, $M_{0001} - M_{1010} = 0.0055 \pm 0.005 \mu_B$, which is nearly completely due to the orbital moment. This is large compared with the anisotropy in the magnetic moment of Ni and Fe, which we calculate to be $M_{001} - M_{111} = 6 \pm 2 \times 10^{-5} \mu_B$ for Ni and for Fe $4 \pm 2 \times 10^{-5} \mu_B$. The order of magnitude is in agreement with experiment [6]. Changing the c/a ratio in cobalt to the experimental value increases E_A by $-1 \mu\text{eV}$, in favour of the c -axis orientation. Under the assumption of the rigid band model the variation of $E_A(n)$ at $n = 9$ for $\delta n = 0.01$ is $2.6 \mu\text{eV/Co atom}$.

$E_A(n)$ for Fe in figure 1c shows less structure than $E_A(n)$ for Ni in figure 1a so that a weaker alloying dependence of the anisotropy constant is expected in Fe, which agrees with experimental observations [6].

In conclusion we believe that magnetic anisotropy energies of the order of $100 \mu\text{eV/Atom}$ or larger can be predicted by *ab initio* calculations; much smaller anisotropies cannot be successfully obtained in this way. Preliminary results for the compounds MnSb and MnAs confirm this picture. Since the calculated anisotropy energy is a difference of ground state energies and our preliminary results for Ni and Fe with the FLAPW method suggest that non-spherical contributions to the potential do not change the magnetic anisotropy energy significantly, we conclude that the difficulty in obtaining small anisotropy energies correctly ($10 \mu\text{eV}$ or less) lies in the local spin density approximation.

We finally mention that a very large anisotropy energy ($300\text{-}400 \mu\text{eV/atom}$) is predicted to occur for a transition metal alloy with 8.7 instead of 9.0 valence electrons in the hcp structure.

- [1] Mori, N. *et al.*, *J. Phys. Soc. Jpn* **37** (1973) 1263; Kondorskii, E. I. and Straube, E., *JETP* **36** (1973) 188; Fritsche, L. *et al.*, *J. Phys. F* **17** (1987) 943.
- [2] Draaisma, H. J. G. *et al.*, *J. Magn. Magn. Mater.* **66** (1987) 351.
- [3] Andersen, O. K., *Phys. Rev. B* **12** (1975) 3060.
- [4] Mackintosh, A. R. and Andersen, O. K., *Electrons at the Fermi Surface*, Ed. M. Springford (CUP) 1980; Weinert, M. *et al.*, *Phys. Rev. B* **32** (1985) 2115.
- [5] Jepsen, O. and Andersen, O. K., *Phys. Rev. B* **29** (1984) 5965.
- [6] Franse, J. J. M., thesis Amsterdam (1969); Paige, D. M. *et al.* (1984) 239; Escudier, P., *Ann. Phys.* **9** (1975) 125.
- [7] Friedel, J., *The Physics of Metals*, Ed. J. M. Ziman (CUP) 1969.
- [8] Wang, C. S. and Callaway, J., *Phys. Rev. B* **9** (1974) 4897.