

# Theory of magnetic interface anisotropy

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Calculations show that the surprising "orientation independence" of the Co/Pd interface anisotropy found experimentally is an accidental consequence of the precise degree of strain exhibited by this system and is not generally expected. The conclusions are supported by the good agreement between experiment and our electronic structure calculations for the anisotropy magnitude, and by the close correlation with the results of a simple symmetry-based theory in which the effects of strain are clear.

The magnetic anisotropy energy determines the direction of magnetization within a uniformly magnetized sample. It is one of the most noticeable and frequently measured features of ferromagnetism. Materials exhibiting itinerant magnetism, such as Fe, Co, and Ni, have attracted particular attention. However, theoretical prediction of the anisotropy energy for these materials poses several difficulties.<sup>1</sup> The anisotropy energy, caused by the spin-orbit interaction, is much smaller than other electronic energies: For the elemental magnets it is, at most, a few  $\mu\text{Ry}$  per atom. This makes predictions sensitive to subtle details of the Fermi surface. Nonelemental samples, such as superlattices, may have larger anisotropies, but comparison to theory is plagued by local variations in layer thickness and other defects. In any case, high numerical accuracy and convergences must be achieved despite the substantial loss of symmetry caused by the spin-orbit interaction. Thus, the computational requirements of a first-principles theory are demanding.

Recently, Engel *et al.*<sup>2</sup> showed that for a system of epitaxially grown Co/Pd superlattices, the interface magnetic anisotropy is positive, large, and orientation independent. Thus, distinctions in anisotropy between superlattices with interfaces in the (111), (100), and (011) directions arise only from magnetostriction and the small bulk Co anisotropy. These results are surprising since the three interfaces involved have very different geometries and atomic areal densities and because previous experiments on Co/Pt and Co/Pd did not display the effect.<sup>2</sup> The origins of the observed independence pose a challenge to the theoretical understanding of anisotropy. In addition, the actual values provide an exceptional quantitative test of theory: they are large, which minimizes numerical problems, and they are derived from a linear superposition of anisotropies taken from a large, but systematic, range of superlattices. This should minimize the problem of samples of multiple composition. A major difficulty, however, is the required calculation of anisotropies for a set of very large superlattices: the experimental Pd thickness corresponds to 4–5 atomic layers (depending on orientation) and the Co thickness ranges to even greater values.

The layer Korringa-Kohn-Rostoker<sup>3</sup> electronic structure method was used to obtain the anisotropy results re-

ported in this paper. This method was chosen because it can efficiently<sup>4</sup> calculate the electronic structure of layered materials, and is not restricted by symmetry to a small subset of superlattices. The spin-orbit interaction was included self-consistently within the scalar relativistic approximation;<sup>5</sup> the details of the implementation will be given in a future publication.<sup>6</sup> The anisotropy was obtained using the force theorem<sup>7</sup> in the following manner: first, a self-consistent electronic structure calculation with a perpendicular spin quantization direction was performed; second, using this self-consistent potential, one iteration for the longitudinal spin quantization direction was performed. The anisotropy energy is given by the difference of the sum of one-electron eigenvalues. A correction of the Fermi energy times the deficit in electrons was added to the eigenvalue sum to account for small deviations from charge neutrality. In the Co/Pd system, we find this perturbation approach yields essentially identical results compared to fully self-consistent total-energy calculations in both directions. This approach differs from that advocated by Daalderop, Kelly, and Schuurmans<sup>1</sup> and Li *et al.*,<sup>8</sup> who both used wave functions from a self-consistent calculation without spin-orbit as a variational basis for a diagonalization of the full Hamiltonian for each spin direction. Demagnetization energies are calculated by summing dipoles as discussed in Ref. 9.

Our results are converged to a numerical accuracy  $\pm 4 \mu\text{Ry}/\text{cell}$  that is roughly 10% of the total magnitude of the anisotropy. This requires 1088 [1620] wave vectors in the two-dimensional Brillouin zone, 48 [48] energy points in the complex integration of the Green's function, and 21 [13] plane waves in the interstitial region for the (100) [(111)] calculation. This corresponds to about 50 000 wave vectors in the three-dimensional zone, a number in agreement with previous calculations.

The positions of our atoms are chosen to be consistent with experimental x-ray-diffraction data.<sup>10,11</sup> The (100) lattice is strained with the lattice constant  $c_{\text{Pd-Pd}} = \sqrt{2}a$  equaling the bulk Pd value 3.89 Å.  $c_{\text{Co-Co}}$  and  $c_{\text{Co-Pd}}$  are 3.29 Å and 3.59 Å, respectively. The (111) lattice is unstrained fcc with lattice constants taken from Ref. 11.

Figure 1 shows the good agreement between experimental and theoretical magnetization<sup>12</sup> for (100) multilay-

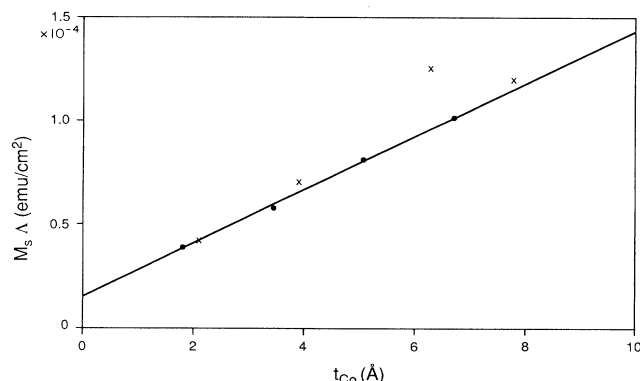


FIG. 1. The Co-thickness dependence of the magnetization multiplied by the superlattice period  $\Lambda$ . Theory is marked by solid circles; experiment (Refs. 2 and 13) is marked by  $\times$ 's. The best linear fit to theory is indicated by the line.

ers, an orientation where the crystal structure is exceptionally well characterized. We have also calculated the anisotropies of the smaller superlattices (100) 1Co/1Pd, (100) 1Co/3Pd, and (111) 1Co/2Pd to allow extensive comparison to earlier linear muffin tin orbital-atomic spheres approximation (LMTO-ASA) results.<sup>14</sup> We find approximate agreement for most superlattices, but do not find their unusually high anisotropy for (100) Co/3Pd. We do, however, find a moderate peak in the anisotropy versus Pd thickness curve at (100) Co/4Pd. This discrepancy may be associated with either the difference in methods or a slight difference in geometry between the two calculations.

The predicted results for anisotropy<sup>12</sup> are displayed in Fig. 2. The interface anisotropies are calculated to be  $0.66 \pm 0.07$  and  $0.57 \pm 0.06$  ergs/cm<sup>2</sup> for the (111) and (100) orientations, respectively. Thus, the interface anisotropy is orientation independent within our numerical accuracy. [The (011) interface anisotropy has not been calculated because this geometry requires many more atoms and plane waves in the interstitial region and, thus, requires additional computational resources.] The experimentally obtained orientation-independent anisotropy

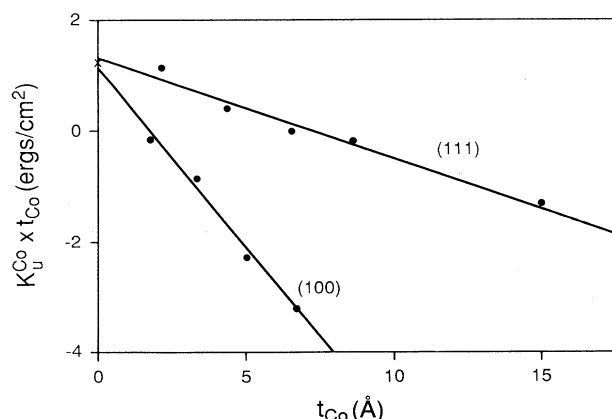


FIG. 2. The theoretical uniaxial anisotropy energy times the Co thickness vs Co thickness. Lines indicate best fit to each orientation. Experimental interface anisotropy is marked by  $\times$ .

was  $0.63 \pm 0.05$  ergs/cm<sup>2</sup>: the comparable theoretical value is  $0.62$  ergs/cm<sup>2</sup>. Thus, theory and experiment agree to within a few percent, which is well within the experimental accuracy. This result strongly suggests that modern electronic structure theory is appropriate for the calculation of large interface anisotropies.

The volume anisotropies are not in such close agreement: the theoretical values are  $-6.4 \times 10^7$  ergs/cm<sup>3</sup> and  $-1.8 \times 10^7$  ergs/cm<sup>3</sup> and the experimental values are  $-4.5 \times 10^7$  ergs/cm<sup>3</sup> and  $-0.5 \times 10^7$  ergs/cm<sup>3</sup> for the (100) and (111) orientations, respectively. Part of this discrepancy is expected. In particular, the (111) (volume anisotropy)/atom (excluding the demagnetization energy) is one order of magnitude smaller than the interface anisotropy and thus may be beyond the accuracy of the electronic structure theory presented here. Furthermore, the experimental sample probably<sup>11</sup> contains some hcp Co (fcc is employed in the calculation), which can bring the two values into much closer agreement. The discrepancy in the (100) volume anisotropy is more substantial: it is possible that this is caused by partial strain relief deep within the Co layer of the experimental sample.

The results for anisotropy may be further analyzed by breaking them out by atom and wave vector. We find that no two-dimensional wave vector dominates the anisotropy: instead, the anisotropy arises from almost all sections of the Brillouin zone. (A contribution from many different wave vectors was also found in the monolayer calculations of Li *et al.*<sup>8</sup>) We also find that the interior Pd atoms contribute little anisotropy. This all suggests that an understanding of the "orientation independence" of interface anisotropy should come from real-space arguments with wave-vector sampling only constituting an averaging factor.

The very deconvolution of the anisotropies into interface and volume contributions suggests that a simple linear model, e.g., the effect of two interfaces is twice that of one interface, is adequate to explain the effect. Symmetry guarantees that the lowest order two atom contribution to the anisotropy is  $L(\mathbf{M} \cdot \mathbf{R})^2$  where  $\mathbf{R}$  connects the two atoms,  $\mathbf{M}$  is the direction of the magnetization, and  $L$  is a constant of unknown sign. Linearity then implies that the interface anisotropy energy may be obtained by summing this basic interaction over all atoms and dividing by interface area. For the special case of the fcc lattice, we find that neglect of all but the nearest-neighbor interactions yields results for the strain and orientation dependence of the anisotropy in excellent agreement with the electronic structure predictions and with a minimum of parameters. This is consistent with the very small value of the more distant hopping integrals, as calculated directly from the electronic structure.

Table I shows results for the interface and volume anisotropies of various strained and unstrained systems. The surface anisotropies may be obtained by setting  $L_m = 0$ : it is clear that the fcc(111), fcc(011), and bcc(111) expressions all differ from those calculated by Néel, who originally postulated this approach.<sup>15</sup> In two of the three cases, this is apparently the consequence of his neglect of

TABLE I. Interface and volume anisotropy energies.  $\theta$  is the angle that the magnetization makes with the interface normal,  $\beta_i$  is the cosine of the angle that the magnetization makes with the vector  $\hat{x}_i$ ,  $a$  is the nearest-neighbor distance in the interface plane,  $L_f$  ( $L_m$ ) is the interaction constant between two ferromagnetic (ferromagnetic and paramagnetic) neighbors, and  $\alpha_f$  ( $\alpha_m$ ) is the cosine of the angle that  $\hat{x}$  makes with the vector connecting ferromagnetic (ferromagnetic and paramagnetic) neighbors on neighboring planes.

System	Strain	Type	Anisotropy energy $\times a^2$
fcc(111)	N	Int.	$[\sqrt{3}(2L_m - L_f)/2] \cos^2 \theta$
fcc(100)	Y	Vol.	$[L_f(6\alpha_f^2 - 3)\sqrt{2 - 2\alpha_f^2}/(a\alpha_f)] \cos^2 \theta$
fcc(100)	Y	Int.	$[2L_m(3\alpha_m^2 - 1) - L_f(3\alpha_f^2 - 1)] \cos^2 \theta$
fcc(100)	N	Int.	$[(2L_m - L_f)/2] \cos^2 \theta$
fcc(011)	Y	Vol.	$L_f(1 - 2\alpha_f^2)(4\beta_y\beta_z - 3\beta_x^2)\alpha_f/(a\sqrt{1 - 3\alpha_f^2}/2)$
fcc(011)	Y	Int.	$[2L_m(6\alpha_m^2 - 3) - L_f(6\alpha_f^2 - 3)]\beta_x^2/2\sqrt{2}$ $+ [2L_m(3 - 4\alpha_m^2) - L_f(3 - 4\alpha_f^2)]\beta_y\beta_z/\sqrt{2}$
fcc(011)	N	Int.	$(2L_m - L_f)\beta_y\beta_z/\sqrt{2}$
bcc(111)	N	Int.	$[\sqrt{3}(2L_m - L_f)/8] \cos^2 \theta$
bcc(100)	N	Int.	0
bcc(011)	N	Int.	$[(2L_m - L_f)/\sqrt{2}]\beta_y\beta_z$
sc(111)	N	Int.	0
sc(100)	N	Int.	$[(2L_m - L_f)/2] \cos^2 \theta$
sc(011)	N	Int.	$-[(2L_m - L_f)/(2\sqrt{2})]\beta_x^2$

one of the two subsurface layers which is connected by first nearest neighbor to the surface atoms.

The value for  $L_f = -60 \mu\text{Ry}$  is obtained by comparing the expression in Table I for the magnetostriction with the volume anisotropy obtained from the (100) electronic structure calculations:  $-5.2 \times 10^7 \text{ ergs/cm}^3$  (excluding the demagnetization energy).  $L_m$  may be obtained by fitting to one or more of the calculated interface anisotropies: all approaches yield a value very close to  $-43.5 \mu\text{Ry}$ . These values yield thickness<sup>12</sup> extrapolated interface anisotropies (includes a small volume contribution) of 0.67, 0.61, and 0.56 ergs/cm<sup>2</sup> for the (111), (011), and (100) orientations, respectively. This indicates that the Co/Pd interface anisotropy is orientation independent to within 7% on the basis of a symmetry argument and a fit to, for example, the (100) data alone. The extent of this orientation independence is very similar to the stated experimental error of 8%.

A key question is whether the approximate orientation independence found experimentally and theoretically is actually weak dependence or is a true independence. We answer this question by examining the unstrained case. Recalculation of the (100) electronic structure results in the absence of strain yields a reduction of the interface anisotropy to  $0.45 \pm 0.06 \text{ ergs/cm}^2$ , thus suggesting that strain was crucial in obtaining the orientation independence. The simple theory, in conjunction with the previously obtained interaction parameters, predicts a similar result:  $0.39 \text{ ergs/cm}^2$ . This suggests that the simple theory accurately represents the effects of strain. However, this theory shows algebraically that true orientation independence only occurs if the strain is fortuitously chosen. Further calculations for Co/Cu and Co/Pt, using a combination of simple theory and electronic structure results, produce substantial orientation dependences,

and thus demonstrate the lack of any general constraint leading to the fortuitous choice of strain. Therefore, the approximate orientation independence found in Co/Pd is technically a weak dependence masked by available accuracies.

The sign of the interface anisotropy is not constrained to be positive by the linear theory. In fact, the theory implies that the unstrained Co surface ( $L_m = 0$ ) should have a negative anisotropy. However, we have searched, via our electronic structure calculations, for interfaces between actual elements that exhibit such an anisotropy: those systems examined [(100) Co/Cu, (111) Fe/Pt, and (111) Co/Pt] all yielded positive values. Furthermore, these results are consistent with several other studies of Co monolayer and interface anisotropies.<sup>14,16</sup> It may be the case that no actual element interacts so weakly with Co that the negative anisotropy criterion of low interaction energy is reached.

In conclusion, the interface anisotropy is shown to be strain and orientation dependent. In the exceptional case of Co/Pd, the precise degree of strain found experimentally masks the orientation dependence to within both the experimental and theoretical accuracy. Particularly strong support for our analysis is provided by the close agreement between experiment, electronic structure calculation, and a simple symmetry-based model for the anisotropy magnitude. This model shows algebraically the relationships between strain, interlayer interactions, and the anisotropy.

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