Magnetic surface anisotropy of cobalt and surface roughness effects within Néel’s model

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Abstract. Néel’s phenomenological model of magnetic surface anisotropy is generalised to hexagonal close-packed structures in order to determine the magnetic surface anisotropy of cobalt. The results obtained are in fair agreement with experimental values for the Au(111)/Co interface. It is also shown that Néel’s model allows the surface roughness to be easily taken into account.

1. Introduction

Theoretical determinations of the magnetic surface anisotropy of transition metals made using band theory are rather difficult because they require an accurate knowledge of the band structure. As a matter of fact, there are only a few such calculations which give more or less reliable results (Bennett and Cooper 1971, Takayama et al 1976, Kolar 1979, Gay and Richter 1986).

Néel (1954) proposed his model in order to introduce the concept of magnetic surface anisotropy but it was 20 years before there was any experimental evidence to support it (Gradmann 1974). Although not very suitable for transition metals, this phenomenological model gives a rather good order of magnitude for the magnetic surface anisotropy of cubic crystals, which is often the only theoretical result to be compared with experiments (Gradmann 1986). However, it sometimes yields the wrong sign for the anisotropy (for example Ni(111)).

The model is extended to hexagonal close-packed structures in § 2 and applied to cobalt in § 3.

Theoretical band calculations of magnetic surface anisotropy generally deal with perfectly flat surfaces. On the other hand, the samples available for experiments always have some surface roughness that will affect the magnetic surface anisotropy. Section 4 is devoted to showing how these roughness effects can be taken into account within Néel’s model.

2. Extension of Néel’s theory to hexagonal close-packed structures

2.1. Presentation of the model

When a cubic crystal is strained or is in a thin-film geometry, the magnetic anisotropy is no longer cubic and lower symmetry terms may appear. The idea of Néel is that the
anisotropies due to magnetostriction and to symmetry breaking at surfaces are connected quantitatively.

In this localised moments model, the magnetocrystalline energy is assumed to be a sum of two-body terms of the form

\[ E_i = g_2(r) P_2(\cos \varphi) + g_4(r) P_4(\cos \varphi) + \ldots \]  

(1)

where \( r \) is the distance between atoms \( i \) and \( j \), and \( \varphi \) is the polar angle between the \( ij \) direction and the common direction of the spins \( S_i \) and \( S_j \), which are bound together by the exchange interaction. \( P_2, P_4, \ldots \) are Legendre polynomials.

It can be shown that the terms \( P_2, P_4, \ldots \) give rise to second- and fourth-order anisotropy terms, which are known to decrease with increasing order. Therefore we shall restrict ourselves to second-order anisotropy and magnetostriction, and neglect all the terms except \( P_2 \).

Now \( g_2(r) \) may be developed in powers of \( \delta r \), where \( \delta r \) is the departure of \( r \) from its equilibrium value. The lowest-order magnetostriction, which is generally sufficient, is obtained by keeping only the first order, i.e. \( g_2(r) = l + m\delta r \).

Since \( g_2(r) \) must decrease faster than \( 1/r^3 \) (Néel 1954) we also make the assumption that only the first neighbours need to be considered. This seems to be a reasonable first approximation.

The important point is that the magnetocrystalline and magnetoelastic effects are described with only two parameters, \( l \) and \( m \). This may not be very accurate but has the enormous advantage of being simple and physically transparent.

2.2. Bulk magnetostriction

First, a theory of magnetostriction is derived that permits the coefficients \( l \) and \( m \) to be obtained from the experimental values of the magnetostriction constants \( \lambda_A, \lambda_B, \lambda_C \) and \( \lambda_D \).

Néel did this calculation for cubic crystals. However, it is more difficult for hexagonal crystals, owing to a more complicated symmetry, and, to our knowledge, has never been done. The tedious detailed calculation is given in the appendix.

Thus the magnetostriction constants are

\[
\lambda_A = \frac{B_1}{2(c_{11} - c_{12})} + \frac{B_2c_{13} - (B_3 + B_1/2)c_{33}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}
\]

\[
\lambda_B = \frac{B_2c_{13} - (B_3 + B_1/2)c_{33}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2} + \frac{B_4}{2(c_{11} - c_{12})}
\]

\[
\lambda_C = \frac{(c_{11} + c_{12}) - c_{13}(2B_3 + B_1)}{2c_{13}^2 - (c_{11} + c_{12})c_{33}}
\]

\[
\lambda_D = \frac{(2B_3 + B_1)(c_{33} - c_{13}) + (c_{11} + c_{12} - 2c_{13})B_2}{4[2c_{13}^2 - (c_{11} + c_{12})c_{33}]} - \frac{B_4}{4c_{44}}
\]

where the \( c_s \) are the usual elastic constants and the \( B_s \) are given by

\[
B_1 = n_\lambda(S_{11} - 2S_{1122}) + n_r m S_{1122}
\]

\[
B_2 = n_\lambda(S_{3333} - S_{33} - S_{1133}) + \frac{1}{2}n_r m (S_{1133} - S_{3333})
\]

\[
B_3 = (n_\lambda - \frac{1}{2}n_r m)(S_{1133} - S_{1122})
\]

\[
B_4 = n_\lambda(S_{11} + S_{33} - 4S_{1133}) + 2n_r m S_{1133}
\]
In formulae (3) \( n \) is the number of atoms per unit volume and the \( \hat{S} \)'s are defined as follows:

\[
\hat{S}_{ijkl} = \sum a_i a_j a_k a_l 
\]

where the \( a_i \)'s are the direction cosines of a neighbouring atom and \( \Sigma \) means a sum over the nearest neighbours.

### 2.3. Anisotropy

The anisotropy energy per atom is

\[
E_a = \frac{1}{2} \sum l(\cos^2\varphi - \frac{1}{3}) = \frac{1}{2} l \sum \beta_i \beta_j \hat{S}_{\varphi}
\]

where the \( \beta \)'s are the direction cosines of the magnetisation. Thus

\[
E_a = \frac{1}{2} l (\hat{S}_{11} - \hat{S}_{33}) \sin^2 \theta
\]

where \( \cos \theta = a_3 \).

For a (0001) surface atom, some neighbours are missing, and this leads to a surface anisotropy energy

\[
E_s = \frac{1}{2} l n_s \sin^2 \theta
\]

where \( n_s \) is the number of atoms per unit area.

For a bulk atom, if one takes \( c/a \) as having the ideal HCP value \( c/a = \sqrt{\frac{8}{5}} = 1.633 \), the bulk anisotropy obtained is zero. This is not due to symmetry, but is rather a numerical coincidence due to the conjunction of taking only nearest neighbours and the ideal ratio \( c/a = \sqrt{\frac{8}{5}} \).

As a matter of fact, if one takes \( c/a = (1 + \delta)\sqrt{\frac{8}{5}} \), in order to take into account the actual ratio \( c/a = 1.622 \) of cobalt, one obtains a non-zero bulk anisotropy:

\[
E_b = -2l \delta n, \sin^2 \theta.
\]

### 3. Magnetic surface anisotropy of (0001) cobalt

The coefficients \( l \) and \( m \) are to be determined from (2) and (3). Since there are four magnetostriction constants \( \lambda_A, \lambda_B, \lambda_C, \lambda_D \) and only two coefficients \( l \) and \( m \), there is generally no solution. This is different from the case of cubic structures where there are only two magnetostriction constants \( \lambda_{(100)} \) and \( \lambda_{(111)} \), which always yield a solution \( (l, m) \).

Thus, one has to solve the system by a least mean squares procedure and take the coefficients \( l, m \) that give the minimum value of \( \Sigma (\lambda'_i - \lambda^0_i)^2 \), where \( \lambda^0_i \) is the actual experimental value and \( \lambda'_i \) is the value computed with \( l \) and \( m \).
Taking the experimental values given in table 1 (Hubert et al. 1969) and table 2 (MacSkimin 1953) one obtains

\[ l = 6.1 \times 10^{-16} \text{erg} \quad r \times m = 1.14 \times 10^{-16} \text{erg} \]

and the \( \lambda \)'s are given in table 3.

Except for \( \lambda_B \), one has good qualitative agreement with the \( \lambda^0 \). This is due to the fact that with only two phenomenological adjustable coefficients, one cannot give an accurate description of the behaviour of the magnetostriction. By taking other neighbours into account and introducing more coefficients, one can nicely fit all the \( \lambda_i \), but one also loses the most important advantage of Néel's model, which is its simplicity.

Finally the anisotropies are

\[ E_b = K^b \sin^2 \theta \quad K^b = 1.2 \times 10^6 \text{erg cm}^{-3} \]
\[ E_s = K^s \sin^2 \theta \quad K^s = 0.84 \text{erg cm}^{-2}. \]

For \( K^b \), the agreement is not very good, since the experimental value at room temperature (the \( \lambda \) and \( c \) values were also taken at room temperature) is \( K^0 = 4 \times 10^6 \) erg cm\(^{-3} \). But this is not very surprising since from (6) \( K^b \) appears as a nearly compensated value. Actually, the anisotropy energy per atom is 100 times smaller for a bulk atom than for a surface atom, owing to the small value of \( \delta \), and one cannot expect to have good agreement for such a small energy. In this case, the nearest-neighbours approximation is no longer valid and second or even third nearest neighbours would contribute to the anisotropy.

On the other hand, the agreement of the magnetic surface anisotropy with experiment is good, both in sign and order of magnitude: for the Au/Co interface Chappert et al. (1986) measured \( K^s = 0.7 \text{erg cm}^{-2} \) at 10 K and \( K^s = 0.5 \text{erg cm}^{-2} \) at 291 K, whereas Bruno et al. (1987) have measured \( K^s = 0.5 \text{erg cm}^{-2} \) at 291 K for the Cu/Co interface.

4. Effect of roughness on the magnetic surface anisotropy

For clarity this section will deal with simple cubic (001) surfaces. The treatment is easily extended to other, more realistic surfaces.

The rough surface is described as follows: it is a flat surface perturbed by square terraces and craters. There are two characteristic parameters: the mean lateral size \( L \)
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of the terraces and craters, and the roughness $\sigma$ which is the mean value of $|\delta z|$ where $\delta z$ is the variation of $z$ from the mean flat surface. $\sigma = |\delta z|$.

For a cubic atom one has

$$E_a = \frac{1}{2} l(S_{11} \beta_1^2 + S_{22} \beta_2^2 + S_{33} \beta_3^2). \quad (9)$$

For a bulk atom $S_{11} = S_{22} = S_{33}$; the anisotropy is zero in this order. For a surface atom $S_{11} = S_{22} \neq S_{33}$, and the energy can be written as $E_a = \frac{1}{2} l(S_{11} - S_{33}) \sin^2 \theta$.

For a step atom $S_{11} \neq S_{22} \neq S_{33}$, but the terraces are square, so the resultant anisotropy is uniaxial and the contribution of an atom can be written as

$$E_a = \frac{1}{2} l[(\frac{1}{2} S_{11} + S_{22}) - S_{33}] \sin^2 \theta. \quad (10)$$

For a surface atom one has $E_a = \frac{1}{2} l \sin^2 \theta$ whereas a step atom gives $E_a = \frac{1}{2} l \sin^2 \theta$. Thus each step atom contributes to a decrease of the anisotropy,

$$\Delta E_a = -\frac{1}{2} l \sin^2 \theta.$$

Hence, the relative decrease in $K_1$ is

$$\Delta K_1/K_1 = -\frac{1}{2} \left( \frac{\text{number of step atoms per unit area}}{\text{number of atoms per unit area}} \right)$$

$$= -\frac{1}{2} \left( \frac{\text{number of terrace (or crater) atoms per unit area}}{\text{number of terrace (or crater) atoms per unit area}} \right) \times \left( \frac{\text{number of terrace (or crater) atoms per unit area}}{\text{number of atoms per unit area}} \right).$$

If the unit length is taken to be the lattice constant, the first and second factors are $4/L$ and $\sigma$ respectively. This gives the following simple, physically meaningful result:

$$\Delta K_1/K_1 = -2\sigma/L. \quad (11)$$

When more realistic surfaces (e.g. FCC (001), FCC (111), HCP (0001)) are investigated the former result remains valid, except for a geometrical factor which depends on the actual shape of the terraces and which is generally close to 1.

It should be emphasised that roughness always reduces the anisotropy, whatever its sign.

This result (11) shows that, for samples prepared under UHV conditions, on good quality substrates, with small roughness and terraces of at least 100 Å (Renard and Nihoul 1987), the roughness effects on anisotropy are negligible. However, when
there is some interdiffusion between the deposit and the substrate, or for sputtered samples, roughness effects may greatly alter the magnetic surface anisotropy.

5. Conclusion

Néel's model of magnetic surface anisotropy has been generalised and applied to HCP (0001) cobalt. The results are in good agreement with experimental results. It has also been shown that one can deal with surface roughness effects within this model.

Therefore, it appears that, whatever its phenomenological character, Néel's model is a useful tool in the study of magnetic surface anisotropies, and it is complementary to band theoretical models rather than in competition with them.

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Appendix. Calculation of the magnetostriction

When a crystal is strained the contributions to the energy are:

(i) the elastic energy:

\[ E_{\text{el}} = \frac{1}{2} c_{11}(\epsilon_{11}^2 + \epsilon_{22}^2) + c_{12} \epsilon_{11} \epsilon_{22} + \frac{1}{2} c_{33} \epsilon_{33}^2 + c_{13}(\epsilon_{11} + \epsilon_{22}) \epsilon_{33} \]

\[ + \frac{1}{2} c_{44}(\epsilon_{13}^2 + \epsilon_{23}^2) + (c_{11} - c_{12}) \epsilon_{12}^2 \]  \hspace{1cm} (A.1)

where \( \epsilon_{ij} \) is the strain tensor and the cs are the elastic constants;

(ii) the magnetoelastic energy, which is for a pair \( i, j \) of atoms

\[ \delta E_{ij} = m \delta r (\cos^2 \varphi - \frac{1}{3}) + l \delta (\cos^2 \varphi). \]

Thus the energy per atom is

\[ w = \frac{1}{2} \sum \left[ m \delta r (\cos^2 \varphi - \frac{1}{3}) + l \delta (\cos^2 \varphi) \right] \]  \hspace{1cm} (A.2)

where \( \sum \) means a sum over the nearest neighbours. \( \delta r \), \( \cos^2 \varphi \) and \( \delta (\cos^2 \varphi) \) can be
expressed as a function of $\varepsilon_{ij}$, the direction cosines $\beta_i$ of the magnetisation and the direction cosines $\alpha_i$ of the neighbour considered:

$$\delta r = r \sum_{i,j} \varepsilon_{ij} \alpha_i \alpha_j$$

$$\cos^2 \varphi = \sum_{ij} \alpha_i \alpha_j \beta_i \beta_j$$  \hfill (A.3)

$$\delta(\cos^2 \varphi) = 2 \sum_{i,j,k} a_i a_k \beta_i \beta_j \varepsilon_{jk} - 2 \sum_{i,j,k,l} a_i a_j a_k \beta_i \beta_j \varepsilon_{jk}$$

then

$$w = \frac{1}{2} \sum \left[ 2l \left( \sum_{i,j,k} a_i a_k \beta_i \beta_j \varepsilon_{jk} - \sum_{i,j,k,l} a_i a_j a_k \beta_i \beta_j \varepsilon_{jk} \right) 
+ m r \left( \sum_{i,j,k,l} a_i a_j a_k \beta_i \beta_j \varepsilon_{jk} - \frac{1}{3} \sum_{i,j} \varepsilon_{ij} \beta_{ij} \right) \right].$$

If one introduces the notation $\delta_{ijkl} = \delta a_i a_j a_k a_l$, it follows that

$$w = l \left( \sum_{i,j,k} \beta_i \beta_j \delta_{ijk} \varepsilon_{jk} - \sum_{i,j,k,l} \beta_i \beta_j \delta_{ijk} \varepsilon_{jk} \right) + m r \left( \sum_{i,j,k,l} \beta_i \beta_j \delta_{ijk} \varepsilon_{jk} - \frac{1}{3} \sum_{i,j} \varepsilon_{ij} \delta_{ij} \right).$$  \hfill (A.4)

From symmetry considerations, many of the $\delta$ are zero, and the only non-zero $\delta$ are

$$\delta_{11} = \delta_{22} = 4 \quad \delta_{33} = 4$$

$$\delta_{1111} = \delta_{2222} = 3 \delta_{1122} = \frac{5}{2} \quad \delta_{2233} = \frac{3}{2} \quad \delta_{3333} = \frac{5}{3}.$$  

It follows that the magnetoelastic energy density can be written as

$$E_{\text{ME}} = B_1 (\beta_1^2 \varepsilon_{11} + 2 \beta_2 \beta_2 \varepsilon_{12} + \beta_3^2 \varepsilon_{22}) + B_2 (1 - \beta_3^2) \varepsilon_{33}$$

$$+ B_3 (1 - \beta_2^2) (\varepsilon_{11} + \varepsilon_{22}) + B_4 (\beta_2 \beta_3 \varepsilon_{23} + \beta_1 \beta_3 \varepsilon_{13}) + B_5 (\varepsilon_{11} + \varepsilon_{22})$$  \hfill (A.5)

with

$$B_1 = n l (\delta_{11} - 2 \delta_{1122}) + n rm \delta_{1122}$$

$$B_2 = n l (\delta_{3333} - \delta_{33} - \delta_{1133}) + \frac{1}{2} n rm (\delta_{1133} - \delta_{3333})$$

$$B_3 = (n l - \frac{1}{2} n rm) (\delta_{1133} - \delta_{1122})$$

$$B_4 = n l (\delta_{11} + \delta_{33} - 4 \delta_{1133}) + 2 n rm \delta_{1133}$$

$$B_5 = - \frac{1}{4} n l (\delta_{1133} + \frac{1}{2} n rm (\delta_{1133} - \delta_{1111})).$$  \hfill (A.6)

Apart from the term $B_4$, which is isotropic in magnetisation and which can be included in the energy of the reference state (demagnetised crystal), this is the magnetoelastic energy given by Mason (1954) from symmetry considerations.
The magnetostriction is obtained by minimising $E_{ME} + E_{EI}$ with respect to $\varepsilon_{ij}$, and one obtains, for the direction $(\gamma_1, \gamma_2, \gamma_3)$,

$$\lambda = \lambda_A [(\beta_1 \gamma_1 + \beta_2 \gamma_2)^2 - (\beta_1 \gamma_1 + \beta_2 \gamma_2)\beta_3 \gamma_3] + \lambda_B [(1 - \beta_3)(1 - \gamma_3) - (\beta_1 \gamma_1 + \beta_2 \gamma_2)^2]$$

$$+ \lambda_C [(1 - \beta_3)\gamma_3^2 - (\beta_1 \gamma_1 + \beta_2 \gamma_2)\beta_3 \gamma_3] + \lambda_D (\beta_1 \gamma_1 + \beta_2 \gamma_2)\beta_3 \gamma_3$$

where $\lambda_A$, $\lambda_B$, $\lambda_C$ and $\lambda_D$, the usual magnetostriction constants (Mason 1954, Hubert et al 1969), are

$$\lambda_A = -\frac{B_1}{2(c_{11} - c_{12})} + \frac{B_2 c_{13} - (B_3 + B_1/2)c_{33}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2}$$

$$\lambda_B = \frac{B_2 c_{13} - (B_3 + B_1/2)c_{33}}{c_{33}(c_{11} + c_{12}) - 2c_{13}^2} + \frac{B_1}{2(c_{11} - c_{12})}$$

$$\lambda_C = \frac{(c_{11} + c_{12})B_3 - c_{13}(2B_3 + B_1)}{2c_{13}^2 - (c_{11} + c_{12})c_{33}}$$

$$\lambda_D = \frac{(2B_3 + B_1)(c_{33} - c_{13}) + B_2(c_{11} + c_{12} - 2c_{13})}{4[2c_{13}^2 - (c_{11} + c_{12})c_{33}]} - \frac{B_4}{4c_{44}}.$$  \hfill (A.7)

From (A.6), (A.7) and the experimental values of the $\lambda$s, one can obtain the coefficients $l$ and $m$.

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