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# Calculation of the magnetic anisotropy energy and finite-temperature magnetic properties of transition-metal films

R Garibay-Alonso<sup>1</sup>, P Villaseñor-González<sup>1</sup>, J Dorantes-Dávila<sup>1,3</sup> and G M Pastor<sup>2</sup>

<sup>1</sup> Instituto de Física, Universidad Autónoma de San Luis Potosí, San Luis Potosí, Mexico

<sup>2</sup> Laboratoire de Physique Quantique, UMR 5626 du CNRS, Université Paul Sabatier, Toulouse, France

E-mail: [jdd@ifisica.uaslp.mx](mailto:jdd@ifisica.uaslp.mx)

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## Abstract

The magnetic anisotropy energy at the interface (IMAE) of Co films deposited on the Pd(111) surface are determined in the framework of a self-consistent, real-space tight-binding method at zero temperature. Significant spin moments are induced at the Pd atoms at the interface which have an important influence on the observed reorientation transitions as a function of Co film thickness. Film–substrate hybridizations are therefore crucial for the magneto-anisotropic behaviour of thin transition-metal films deposited on metallic non-magnetic substrates. Furthermore, using a real-space recursive expansion of the local Green function and within the virtual-crystal approximation we calculate the magnetization curves and the Curie temperature  $T_C$  for free-standing Fe films.

## 1. Introduction

Magnetic nanostructures involving transition metals (TMs) is a very active field mainly for the possibility of tailoring new materials with specific applications [1]. In particular, the study of reorientation transitions of ultra-thin films induced either by modifying the interfaces of magnetic materials, for example by capping thin magnetic films with non-magnetic elements [2–5], or by finite-temperature effects [6, 7] is a subject of central interest from both fundamental [8] and technological standpoints. The control and understanding of these phenomena at an atomic level are central issues in current research on magnetism with considerable implications for the development of magnetic nanostructures at surfaces.

Concerning finite-temperature effects, most of the calculations of the magnetic properties have been quite successful in predicting the experimental behaviour of bulk particularly

<sup>3</sup> Author to whom any correspondence should be addressed.

at low temperatures [9, 10]. In contrast, very little is still known about the theory of TM low-dimensional magnetism at finite temperatures. In order to derive reliable conclusions concerning the local environment dependence of the magnetization curves and Curie temperatures ( $T_C$ ), the electronic theory must take into account both the fluctuations of the magnetic moments and the itinerant character of the d-electron states.

In this paper, first we present results on the magnetic anisotropy at the interfaces of capped magnetic Co thin films on Pd(111) by changing the capping element, then we present a functional integral theory for low-dimensional systems taking the Fe free-standing films as a representative example of transition metals.

## 2. Co films on Pd(111) at $T = 0$

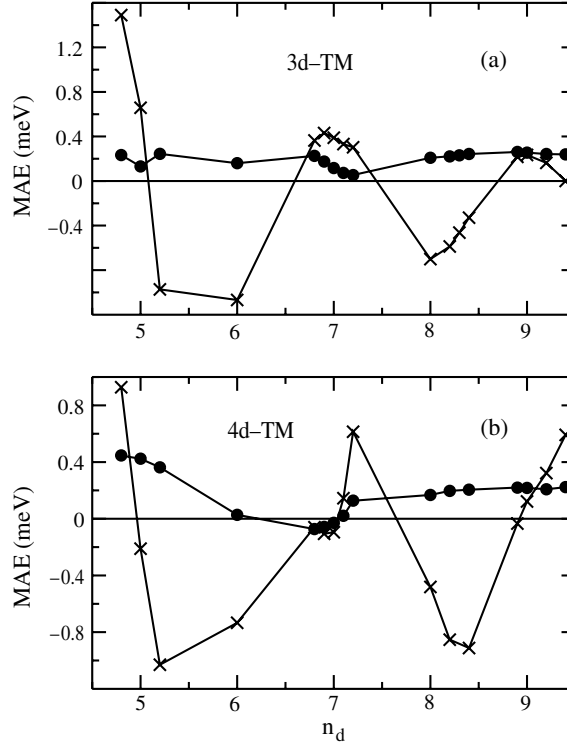
A particularly relevant example of tailoring magnetic behaviour at a nanoscopic scale is the experimental observation that magnetization reorientation transitions can be induced by modifying the interfaces of magnetic materials, for example by capping thin magnetic films with non-magnetic elements [3–5]. Measurements on Co films deposited on Pd(111) show that the easy axis of uncovered  $\text{Co}_m/\text{Pd}(111)$  is within the film plane, while capped  $\text{Pd}_n/\text{Co}_m/\text{Pd}(111)$  already presents an off-plane easy axis for  $n \geq 1$  ( $m \leq 8$ ) [3].

Layer-resolved self-consistent electronic calculations of magnetic anisotropy energy (MAE) [8] provide new insight into the capping-induced off-plane magnetization observed in Co films on Pd(111). These reorientation transitions, before and after capping and as a function of  $m$ , are in good agreement with torsion–oscillation magnetometry experiments [3]. Moreover, it has been shown that the transition from perpendicular to in-plane phases with increasing film thickness involves an intermediate canted phase in qualitative agreement with recent experiments [5].

In addition, the interfaces responsible for the stability of the off-plane easy axes are characterized microscopically. For example, results for the layer-resolved contributions of the magneto-crystalline anisotropy energy  $\Delta E^{\text{MC}}(l)$  and for the interface magnetic anisotropy energy (IMAE)  $\Delta E_1$  given by the sum of  $\Delta E_{zx}^{\text{MC}}(l)$  for the two adjacent Co and Pd layers reveal an unexpected internal magnetic structure of the Co–Pd interfaces. Even–odd oscillations of  $\Delta E_{zx}^{\text{MC}}(l)$  for small  $m$  are present. However, it is remarkable that, despite such a strong  $m$  dependence of  $\Delta E_{zx}^{\text{MC}}(l)$ , the Co–Pd substrate IMAE remains approximately independent of  $m$  ( $m \geq 2$ ), and is not much affected by the presence of capping, even for the smallest  $m$  [8].

The overall MAE of Pd-capped Co films shows a qualitatively different behaviour. For a single layer of Pd capping ( $n = 1$ ) the Co monolayer ( $m = 1$ ) shows a perpendicular magnetization with a rather large  $\Delta E_{zx} = 0.4$  meV. As  $m$  increases,  $\Delta E_{zx}$  increases mainly due to the growing importance of the dipole–dipole contribution  $\Delta E_{zx}^{\text{DD}} < 0$ . Therefore, for thick enough films  $\Delta E_{zx}^{\text{DD}}$  dominates over  $\Delta E_{zx}^{\text{MC}}$  and  $\vec{M}$  turns back to in plane for  $m = 7$ –8.

Furthermore, it is also of considerable interest to assess the possibility of tailoring IMAE by changing the capping material. In order to investigate the specificity of Pd as capping element, and the possible effect of sp–d charge transfers, we have performed calculations of the MAE of a capped three-layer Co film  $\text{X}_1/\text{Co}_3/\text{Pd}(111)$  as a function of the d-band filling  $n_d$  of the capping transition metal X. For the calculations, we consider a self-consistent tight-binding method developed in [8] and [11]. The d-electron exchange integral and hopping elements of Co (Pd) are used for the overlayer as representative values for 3d (4d) TMs. In figure 1 results are given for the IMAE as a function of  $n_d$ . The IMAE formed by the Pd layer substrate and the first layer of the Co film is always positive and in some cases close to zero (dots in the figure). In contrast, we obtain that the  $\Delta E_1$  formed by the capping element and the Co surface layer oscillates as a function of  $n_d$ , being positive around  $n_d = 5, 7$  and 9



**Figure 1.** Interface MAE  $\Delta E_1$  in  $\text{TM}_1/\text{Co}_3/\text{Pd}(111)$  films as a function of the d-band filling  $n_d$  of the capping (a) 3d and (b) 4d transition metal (TM). Results are given for  $\Delta E_1 = \Delta E_{\text{Co}}^{\text{MC}}(3) + \Delta E_{\text{Pd}}^{\text{MC}}(4)$  at the Co–Pd interface (dots) and for  $\Delta E_1 = \Delta E_{\text{Co}}^{\text{MC}}(1) + \Delta E_{\text{TM}}^{\text{MC}}(0)$  at the TM–Co capping interface (crosses).

and negative around  $n_d = 6$  and  $8$  (see crosses in the figure). The quantitative importance of  $\Delta E_1$  and the precise ranges of  $n_d$  for positive and negative values depend on whether 3d or 4d TMs are considered, mainly due to the different ratio  $J/W$  between the d-electron exchange integral and band width. Rather large negative  $\Delta E_1$  are obtained for Co ( $n_d \sim 8$ ), Rh ( $n_d \sim 8$ ) and Nb ( $n_d \sim 5$ – $6$ ). In contrast, Cr ( $n_d \sim 5$ ), Fe ( $n_d \sim 7$ ) and Pd ( $n_d \sim 9.5$ ) capping favours off-plane magnetization, while for Ni ( $n_d \sim 9$ ) and Ru ( $n_d \sim 7$ )  $\Delta E_1$  is smaller and the tendency is less clear. Large  $\Delta E \sim 0.5$  meV are also obtained for Cr, in good agreement with the recent observation of perpendicular magnetization in Cr/Co<sub>m</sub>/Pd(111) films [4]. Changing the capping material offers certainly interesting possibilities of tailoring the interface MAE.

### 3. Magnetization curves of Fe films

The finite-temperature magnetic properties of thin films are determined by applying the functional-integral formalism developed by Hubbard and Hasegawa for TM solids [9]. The many-body interaction  $H_1$  is written as

$$H_1 = \frac{1}{2} \sum_i \left[ (U - J/2) \hat{N}_i^2 - 2J \hat{S}_{iz}^2 - (U - J) \hat{N}_i \right], \quad (1)$$

where  $\hat{N}_i = \sum_{\alpha\sigma} \hat{n}_{i\alpha\sigma}$  is the number of electron operator at atom  $i$ , and  $\hat{S}_{iz} = (1/2) \sum_{\alpha} (\hat{n}_{i\alpha\uparrow} - \hat{n}_{i\alpha\downarrow})$  is the  $z$  component of the spin operator.  $U = (U_{\uparrow\downarrow} + U_{\uparrow\uparrow})/2$  and  $J = U_{\uparrow\downarrow} - U_{\uparrow\uparrow}$

stand for the average direct and exchange Coulomb integrals which are taken to be orbital independent. For the calculation of the partition function  $Z$ , the quadratic terms in equation (1) are linearized by means of a two-field Hubbard–Stratonovich transformation within the static approximation. A charge field  $\eta_i$  and an exchange field  $\xi_i$  are thus introduced at each site  $i$ . These represent the local finite-temperature fluctuations of the d-electron energy levels and exchange splittings, respectively. Using the notation  $\vec{\xi} = (\xi_1, \dots, \xi_N)$  and  $\vec{\eta} = (\eta_1, \dots, \eta_N)$ ,  $Z$  is given by

$$Z \propto \int d\vec{\eta} d\vec{\xi} \tilde{Z}(\vec{\xi}, \vec{\eta}), \quad (2)$$

where

$$\tilde{Z}(\vec{\xi}, \vec{\eta}) = \exp\{-\beta F(\vec{\xi}, \vec{\eta})\} \quad (3)$$

$$= \exp\left\{-\frac{\beta}{2} \sum_i \left[ \left( U - \frac{J}{2} \right) \eta_i^2 + \frac{J}{2} \xi_i^2 \right] \right\} \text{Tr}[\exp\{-\beta(\hat{H}_{\text{eff}} - \mu \hat{N})\}]. \quad (4)$$

$\hat{H}_{\text{eff}}$  describes the dynamics of the d electrons as if they were independent particles moving in a random alloy with energy levels  $\tilde{\varepsilon}_{i\sigma}$  given by

$$\tilde{\varepsilon}_{i\sigma} = \varepsilon_i^0 + \left( U - \frac{J}{2} \right) i\eta_i - \sigma \frac{J}{2} \xi_i. \quad (5)$$

The thermodynamic properties of the system are obtained as a statistical average over all possible distributions of the energy levels  $\tilde{\varepsilon}_{i\sigma}$  throughout the system.

Since we are mainly interested in the magnetic properties and since  $J \ll U$ , we neglect the thermal fluctuations of the charge fields  $\eta_i$  by setting them equal to their exchange-field-dependent saddle-point values  $i\vec{\eta}_i = \langle \hat{N}_i \rangle$ . This amounts to a self-consistent determination of the charge distribution for each exchange field configuration  $\vec{\xi}$ . In this way

$$Z \propto \int d\vec{\xi} \tilde{Z}(\vec{\xi}), \quad (6)$$

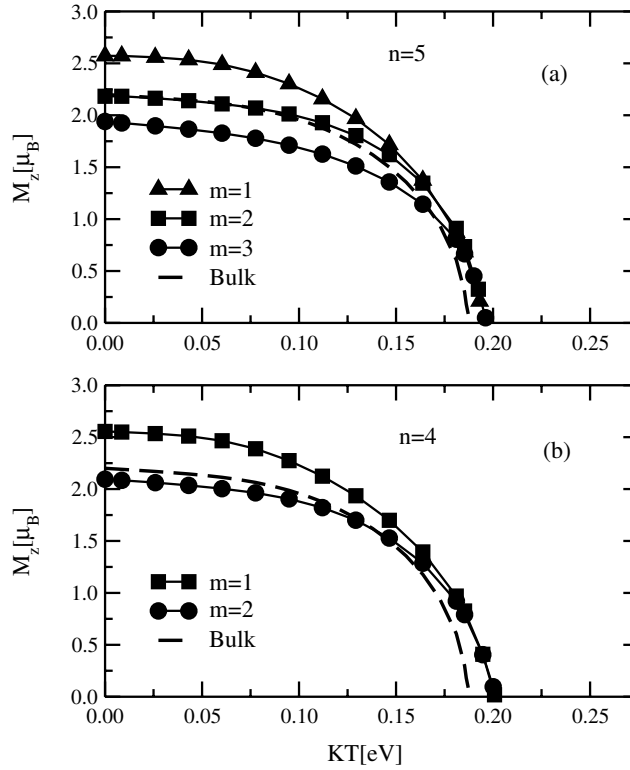
where  $\tilde{Z}(\vec{\xi})$  depends now only on the most relevant exchange variables  $\xi_i$  that describe the fluctuations of the spin degrees of freedom.

The integrand  $\exp\{-\beta F(\vec{\xi})\}$  is interpreted as proportional to the probability  $P(\vec{\xi})$  for a given exchange-field configuration  $\vec{\xi}$ . The thermodynamic properties are obtained by averaging over all possible  $\vec{\xi}$  with  $\exp\{-\beta F(\vec{\xi})\}$  as weighting factor. For example, the local magnetization at atom  $i$  is given by the average of  $2\langle S_{iz} \rangle = \sum_\alpha \langle \hat{n}_{i\alpha\uparrow} - \hat{n}_{i\alpha\downarrow} \rangle$  which depends on  $\vec{\xi}$  and fluctuates at  $T > 0$ :

$$\mu_i(T) = \int \xi_i P_i(\vec{\xi}) d\vec{\xi}. \quad (7)$$

Here,  $P_i(\vec{\xi}) = \frac{1}{Z} \exp\{-\beta F_i(\vec{\xi})\} = \frac{1}{Z} \int \prod_{l \neq i} d\xi_l \exp\{-\beta F(\xi_1, \dots, \xi_{i-1}, \xi, \xi_{i+1}, \dots, \xi_N)\}$ . Thus, the temperature-dependent local magnetization is equal to the average of the local exchange field. Equation (7) justifies the intuitive association between the fluctuations of the local moment  $2\langle \hat{S}_{iz} \rangle$  at atom  $i$  and those of the exchange field  $\xi_i$ .

Equation (7) suggests that the problem of determining the magnetic properties of Fe thin films at finite temperature can be solved in a similar way as that of random alloys. Thus, in this context, a given configuration of the exchange fields  $\vec{\xi}$  occurs with a probability  $P(\vec{\xi})$ . A first insight into the magnetic behaviour of Fe thin films can be obtained by treating the disorder within the virtual crystal approximation (VCA) [12]. More sophisticated approximations like the coherent potential approximation (CPA) are also likely to be used.



**Figure 2.** Temperature dependence of the magnetization for a  $n$ -layer Fe film: (a) corresponds to results for a five-layer film and (b) for a four-layer film. For  $n = 5$  and  $4$ ,  $m = 1$  refers to the surface layer and  $m = 2$  to the layer below the surface. For  $n = 5$ ,  $m = 3$  refers to the central layer. Dashed curves show the bulk results.

The parameters used for the calculations on  $\text{Fe}_N$  are the same as in [13], namely, bulk  $d$ -band width  $W = 6.0$  eV, direct Coulomb integral  $U = 6.0$  eV and exchange integral  $J = 0.70$  eV. The electronic properties are obtained from the local densities of states (DOSs) which are computed by using the Haydock–Heine–Kelly recursion method.

The results for the temperature dependence of the magnetization [ $M(T)$ ] of  $n$ -layer Fe films oriented in the (001) direction are shown in figure 2. For the bulk (dashed curves), we obtain  $T_C \simeq 1900$  K ( $T_C^{\text{exp}} = 1043$  K). A similar result was obtained recently by using *ab initio* calculations and dynamical field theory [10]. However, the behaviour of  $M(T)$  close to  $T_C$  is different from that experimentally obtained (see the sharp transition in figure 2 at  $T = T_C$ ). In fact, the VCA approximation has serious problems for alloys with similar concentrations, i.e., close to 0.5 (in our case, negative and positive fluctuations  $\xi^-$  and  $\xi^+$  with similar probabilities). However, preliminary results obtained by using more sophisticated approximations such as the CPA show that the observed main trends of  $M(T)$  as a function of system size are similar to those obtained by using the VCA.

In addition to the bulk  $M(T)$  we show the results for the (001) five-layer (figure 2(a)) and four-layer (figure 2(b)) Fe film. Notice that for  $n = 4, 5$   $T_C$  is slightly larger as compared to that of the bulk ( $T_C(n = 4, 5) \simeq 2000$  °C). This is due to the higher local magnetic moment increases as compared to the bulk one. In this case, the large value of the exchange splitting dominates the reduced dimensionality (smaller coordination number). This shows the subtle

electronic behaviour at finite temperatures of transition-metal systems. We observe that the behaviour of  $M(T)$  for the central layer is similar to that of the bulk.

In conclusion, we have obtained the magnetization curves of bulk Fe and of the  $n$ -layer Fe film with the (001) surface orientation. We have shown that the electronic structure effects due to the itinerant character of the d electrons are very important in the determination of the magnetic behaviour of low-dimensional systems at finite temperatures. While these results are valuable, several important aspects of the problem still remain to be addressed. First, it would be interesting to investigate to what extent our calculations are influenced by certain desirable improvements. In fact, the VCA is a good approximation for the low-temperature limit, where all but a few fields  $\xi_i$  are kept equal to the  $T = 0$  result. This is not the case for  $T \simeq T_C$  in which this approximation fails. To improve our results, for example, one could treat disorder by using the CPA. This could certainly modify the magnetic behaviour close to  $T_C$ . Substrate effects are also desirable to be investigated. These investigations are currently in progress.

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