THEORY OF THE CURIE TEMPERATURE OF COBALT-BASED FERROMAGNETIC ULTRATHIN FILMS AND MULTILAYERS (Invited)

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Abstract—Calculations of the Curie temperature and magnetization in cobalt-based ultrathin films and multilayers have been performed within the framework of the Green-function theory. The effect of the long-ranged dipolar interactions and of the strong interface magnetic anisotropy is fully taken into account. The influence of the interlayer coupling across the non magnetic spacer is also investigated. The results are discussed in connection with experimental data for Co/Au, Co/Pt, and Co/Pd multilayers.

KEYWORDS: MULTILAYERS, CURIE TEMPERATURE, GREEN FUNCTION, MAGNETIC ANISOTROPY, DIPOLE INTERACTIONS, INTERLAYER COUPLING

INTRODUCTION

In the last few years, ferromagnetic ultrathin films have been shown to exhibit remarkable interface-induced properties. In particular strongly enhanced magnetic anisotropies [1] and magneto-optical effects [2] have been found in Co/Au, Co/Pd, and Co/Pt ultrathin films and multilayers. Cobalt-based multilayers with extremely low individual cobalt thicknesses (a few atomic layers) thus present a strong perpendicular anisotropy, together with a large magneto-optical Kerr or Faraday rotation; these properties are precisely the two major requirements for magneto-optical storage technology. Indeed cobalt-based multilayers have been proved to be very promising candidates as high-density magneto-optical storage materials [3-6]. They present over the traditional rare-earth-transition-metal alloys the advantages of a higher chemical stability, together with larger magneto-optical effects at small wavelengths, thus allowing higher storage densities.

The stability of the written information and the thermomagnetic writing process used in magneto-optical technology impose severe requirements on the Curie temperature: an optimal value for the Curie temperature is in the range of 400-600 K [7]. In rare-earth-transition-metal alloys, the Curie temperature can be tuned by varying the alloy composition. The Curie temperature of bulk cobalt is much too high for this purpose; fortunately it is much smaller in ultrathin films, and strongly thickness dependent. It can therefore be

adjusted to a desired value by a proper choice of the cobalt thickness.

In bulk materials, the Curie temperature is determined essentially by the magnitude of the exchange interaction. Dipole interactions and magnetic anisotropy, which are much weaker, contribute to small corrections and are therefore negligible in a first approximation. The situation is completely different in the case of ultrathin films and multilayers. Indeed, in the absence of dipole interactions and magnetic anisotropy, these quasi two-dimensional systems should have a zero Curie temperature, according to the Mermin-Wagner theorem [8].

In Co/Pd multilayers—and to a less extent in Co/Pt multilayers—the strong exchange-enhanced susceptibility of the spacer layer (Pd) induces a ferromagnetic coupling between the individual cobalt layers [9]. This coupling is expected to influence the Curie temperature, and is therefore to be taken into account.

We have used the Green-function formalism within the random phase approximation [10] to calculate the Curie temperature in cobalt-based multilayers. A generalization of the existing formalism for ultrathin films [11] has been developed in order to take fully into account the long-ranged dipole interactions and the magnetic anisotropy [12].

The paper is organised as follows. The model is presented in Sec. 1. The results for the Curie temperature of ultrathin films and multilayers are given in Sec. 2. The results are discussed in con-

nection with available experimental data in Sec. 3.

MODEL

The cobalt ultrathin film is assumed to have a hcp structure, with the (0001) axis normal to the film plane. The spins are assumed to be localized, and have the same magnitude as in bulk cobalt (i.e. $S = 0.86$). The hypothesis of localized magnetism is in principle not very good for a transition metal; however, we can hope that itinerant-magnetism features like Stoner excitations will not be very important with respect to the Curie temperature, which is mainly determined by the low-lying excitations (the spin waves).

The exchange interaction is represented by a Heisenberg Hamiltonian, with the same exchange integral J as in bulk cobalt,

$$\mathcal{H}_{\text{ex}} = - \sum_{\langle m_i, n_j \rangle} J \vec{S}_{m_i} \cdot \vec{S}_{n_j}. \quad (1)$$

In the above equation, the indices m and n refer to the atomic planes, whereas the i and j label the atomic positions within the planes, and the notation $\langle m_i, n_j \rangle$ indicates that the sum is restricted to nearest neighbors pairs. The dipole-interaction Hamiltonian is given by the usual expression

$$\mathcal{H}_{\text{dip}} = \frac{1}{2} (2\mu_B)^2 \sum_{m_i, n_j} \frac{1}{r_{m_i, n_j}^3} \left[\vec{S}_{m_i} \cdot \vec{S}_{n_j} - 3 \left(\vec{S}_{m_i} \cdot \vec{u}_{m_i, n_j} \right) \left(\vec{S}_{n_j} \cdot \vec{u}_{m_i, n_j} \right) \right]. \quad (2)$$

The magnetic anisotropy is introduced as a pseudo-dipolar interaction between nearest neighbors,

$$\mathcal{H}_K = \frac{1}{2} \sum_{\langle m_i, n_j \rangle} K_{m, n} \left[\vec{S}_{m_i} \cdot \vec{S}_{n_j} - 3 \left(\vec{S}_{m_i} \cdot \vec{u}_{m_i, n_j} \right) \left(\vec{S}_{n_j} \cdot \vec{u}_{m_i, n_j} \right) \right]. \quad (3)$$

The interaction constants for neighbors pairs located respectively in the same atomic layer ($K_{m, m}$) and in neighboring atomic layers ($K_{m, m \pm 1}$) are chosen in order to yield the volume and surface anisotropy constants typically observed in hcp (0001) cobalt ultrathin films [1]. The total

anisotropy field (including the dipolar contribution) can finally be written

$$H_K^{\text{eff}} = H_K^V + \frac{2H_K^S}{N} - 4\pi M_0, \quad (4)$$

where N is the number of atomic planes, $H_K^V = 6$ kOe the volume anisotropy field, $H_K^S = 40$ kOe the interface anisotropy field, and $4\pi M_0 = 18.2$ kOe the dipolar demagnetizing field. One thus obtains a perpendicular anisotropy for cobalt thicknesses below 7 atomic layers, as depicted in Fig. (1). The interlayer coupling can be characterized by the *interlayer-coupling field*, defined by $H_i = A(M_0 d)^{-1}$, where A in erg.cm^{-2} is the interlayer exchange constant, and d the thickness of one atomic layer.

Fig. 1. Product $N H_K^{\text{eff}}$ as a function of N .

The random-phase-approximation-Green-function formalism used here to compute the Curie temperature is an extension of previous theories for three-dimensional systems [10] and ultrathin films [11], and will be published elsewhere [12].

RESULTS

The calculated values for the Curie temperature of ultrathin hcp (0001) cobalt films are shown

on Fig. (2) (solid circles); the other points refer to experimental data and will be discussed below. These results are also relevant for uncoupled multilayers. As expected, we observe that the Curie temperature rapidly increases with the cobalt thickness. From these calculations, the optimal cobalt thickness for magneto-optical applications lies between 1 and 2 atomic layers.

Fig. 2. Curie temperature of ultrathin hcp (0001) cobalt films, as a function of the number of atomic planes N . Calculation results (solid circles), and experimental results for Co/Pt multilayers from Ref. [5] (triangle) and from Ref. [13] (diamonds)

The coupling strength of a Pd spacer has been measured by Celinski *et al.* [9]. It presents approximately a decreasing exponential thickness-dependence; extrapolating their results down to 1 atomic layer yields an interlayer-coupling field of 100 kOe. The calculated Curie temperature of coupled bilayers and multilayers are shown respectively in Figs. (3) and (4). The increase of the Curie temperature with the interlayer coupling H_i and with the number P of periods is well demonstrated.

DISCUSSION

Experimental results for the Curie temperature of Co/Pt multilayers prepared by electron-beam evaporation [5, 13] are displayed in Fig. (2).

Such systems are more likely to present sharp interfaces than sputtering-prepared multilayers. The experimental data are seen to agree quite well with calculated results for uncoupled multilayers. However, a larger set of experimental results would be desired for a better comparison.

Fig. 3. Calculated Curie temperature of coupled bilayers as a function of the coupling field H_i , for various thicknesses N of the individual Co layers

A striking feature of the experimental results is the strong variation of the Curie temperature of Co/Pt multilayers with respect to the platinum thickness: the upmost and lowest diamonds in Fig. 1 correspond to platinum thicknesses of respectively 4.5 and 9.5 atomic layers [5]. An even stronger influence of the platinum thickness is reported for sputtered Co/Pt multilayers [14]: for a cobalt thickness of approximately 2 atomic layers, the Curie temperature drops from 425 K to 200 K, when the platinum thickness is increased from 3 to 9 atomic layers. Can such an effect be explained by the interlayer coupling across the platinum layer? This is very unlikely. Actually, if we try to evaluate the magnitude of the coupling for such Co/Pt multilayers from the measurements of Ref. [9], taking into account the fact that the susceptibility of platinum is one order of magnitude smaller than in palladium, we obtain that the interlayer coupling field

for 3 atomic layers of platinum is of the order of magnitude of 4 kOe. As can be seen from Figs. (3) and (4), such a coupling cannot explain the strong platinum-thickness dependence of the Curie temperature reported in Refs. [5, 14]. Rather this could be due to interdiffusion between Co and Pt, which are known to form solid solutions. This interpretation is supported by a comparative study of Co/Au and Co/Pt multilayers by Bloemen *et al.* [15].

Fig. 4. Calculated Curie temperature of coupled multilayers as a function of the number of periods, for a coupling field of 100 kOe

Stronger coupling effects can be expected in Co/Pd, due to the very high susceptibility of palladium. Unfortunately, experimental data for the Curie temperature of Co/Pd multilayers are still lacking.

CONCLUSION

The Curie temperature of cobalt-based multilayers for magneto-optical storage has been investigated theoretically. The Curie temperature can be adjusted to the desired value by varying the cobalt thickness. The optimal cobalt thickness is found to lie between one and two atomic layers. The interlayer coupling across a platinum spacer is found to have only a minor influence on the Curie temperature. The observed influence of the platinum

thickness on the Curie temperature could be due to some interdiffusion of platinum and cobalt.

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