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Perpendicular anisotropy in Pd/Co multilayers

H. J. G. Draaisma

Department of Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

F. J. A. den Broeder

Philips Research Laboratories, 5600 JA Eindhoven, The Netherlands

W. J. M. de Jonge

Department of Physics, Eindhoven University of Technology, 5600 MB Eindhoven, The Netherlands

The anisotropy values in polycrystalline Pd/Co multilayers with [111] texture as determined from magnetization measurements, torque measurements, and ferromagnetic resonance are compared. It is concluded that the most reliable value is obtained from the area between the magnetization curves measured with the field parallel and perpendicular to the plane of the film. The anisotropy is made up of a volume and an interface contribution. In a pair interaction model for the interface anisotropy it is shown that the latter is very sensitive to the precise distribution of Co and Pd at the interfaces. From the experimental data we derive a lower bound value of 0.2 meV per Co atom for the energy change with the direction of the magnetic moment. The temperature dependence of the magnetization indicates a long-range interaction across the Pd layers up to 15–25 Å.

I. INTRODUCTION

One of the intriguing aspects of magnetic multilayers is the reduced symmetry at the interfaces, which can lead to a considerable contribution to the total anisotropy. In polycrystalline Pd/Co multilayers with [111] texture this leads to a preferred direction for the magnetization perpendicular to the film plane when the Co thickness is below 8 Å.^{1–3} The values for the anisotropy in these multilayers were obtained from the area between the parallel and perpendicular magnetization curves as measured in a vibrating sample magnetometer (VSM). As was shown, the anisotropy was independent of the thickness of the Pd layers and could be interpreted as the sum of an interface and a volume contribution of the Co layers. The influence of the Pd layers on the magnetization curves was described in a separate paper.⁴ In this paper we present additional measurements on new samples of Pd/Co, prepared by electron-beam evaporation, and compare the values for the total anisotropy resulting from torque and magnetization measurements with the values of previous samples¹ and of Pd/Co multilayers prepared by rf sputtering.³ Further we examine the effect of interdiffusion on the anisotropy with a pair interaction model and the temperature dependence of the magnetization.

II. TORQUE MEASUREMENTS

Torque measurements provide a direct way to determine the magnetic anisotropy in ferromagnetic materials.⁵ In thin films with rotational symmetry around the axis perpendicular to the film plane, the anisotropy can be written as

$$K(\Theta) = K_1 \sin^2 \Theta + K_2 \sin^4 \Theta + \dots, \quad (1)$$

in which Θ is the angle between the magnetization and the perpendicular axis. K_1 includes the magnetostatic energy ("demagnetization") as a negative term, since in multilayers with inhomogeneous magnetization this contribution can not be distinguished from contributions of crystalline

and magnetoelastic origin with the same angular dependence. The most accurate way to interpret the torque T as a function of the angle φ between the field and the axis normal to the film plane, is to determine the slope of the curve at angles where the torque is zero.⁶ It can easily be found that for $\varphi = 0$ this slope can be written as

$$\left(\frac{dT}{d\varphi}\right)_{\varphi=0} = -\left(\frac{1}{2K_1} + \frac{1}{MB_0}\right)^{-1}, \quad (2)$$

in which B_0 is the applied magnetic field (in T) and M is the magnetization in the film (in A/m). B_0 should be large enough to saturate the sample and keep the angle between the magnetization and the applied field small. When K_1 is positive, the slope will be negative and vice versa.

For a series of Pd/Co multilayers with varying Co thickness we measured the torque curve using a standard torque magnetometer (TRT-2 from Toei Kogyo Co., Japan) in a field $B_0 = 1.75$ T, which was the highest field available. The results, shown in Fig. 1, indicate a sign change of the anisotropy for Co layers between 6 and 10 Å in agreement with previous VSM measurements.¹ In Fig. 2 the values K_1 found from the slope of the torque curves are compared to the values determined from the area between the magnetization curves with the magnetic field parallel and perpendicular to the film plane. Though the last method measures the total anisotropy $K = K_1 + K_2 + \dots$, K_1 is often the dominating term. For the samples with Co thickness larger than 6 Å the comparison is quite good, but deviations begin to occur for 4 Å. In the case of 2 Å Co the values differ by a factor of 3. From the magnetization curves we know that 1.75 T is not enough to saturate these samples in other than the perpendicular direction, but if that would be the cause of the deviation we would expect a lower value from the torque measurement instead of a higher. It is possible however, that the large hysteresis ($\mu_0 H_c = 0.3$ T) in the multilayers with 2 Å Co disturbs the measurement and that higher fields are necessary.

FIG. 1. Torque per volume Co as function of the angle φ between the applied field ($B_0 = 1.75$ T) and the axis perpendicular to the film plane indicates a sign change of the anisotropy as the Co layers get thinner. From the slope of the tangent at $\varphi = 0$, given by the straight lines, the anisotropy constant K_i can be calculated.

The anisotropy can also be determined from the uniform resonance mode in a ferromagnetic resonance experiment (FMR).⁷ Again the values found for Co thicker than 6 Å coincide with the values in Fig. 2, but for the thinner Co layers domain effects are observed, which prevent the determination of the anisotropy.⁸

Therefore, we use the values from the VSM measurement to estimate the interface and volume contribution to the anisotropy. For the present samples we find $K_s = 0.55 \times 10^{-3}$ J/m² and $K_v = 1.2 \times 10^6$ J/m³. Previously we found $K_s = 0.26 \times 10^{-3}$ J/m² and $K_v = -0.72 \times 10^6$ J/m³,¹ while for rf-sputtered multilayers $K_s = 0.16 \times 10^{-3}$ J/m² and $K_v = -0.37 \times 10^6$ J/m³ were found.³ It is to be noted that these figures result from a series of samples, which means that the deviations are caused by systematic differences between the series. The present samples were deposited onto a 100 Å Ti layer to improve the sticking to the glass substrate, but this did not seem to affect the structural characteristics as measured by x-ray diffraction. Two things are remarkable: First, the experimental interface anisotropy is

FIG. 2. Anisotropy per area of one Co layer as function of the thickness of the Co layers reveals the interface and volume contribution to the anisotropy. A comparison is made for rf-sputtered samples (Ref. 3), previously vapor-deposited samples (Ref. 1), and the present samples, also prepared by vapor deposition. The data are given for these latter samples. The length of the lines indicates the range of Co thicknesses studied in each series of experiments.

not a fixed quantity, but seems to fall in a wide range of possible values. Second, when the interface contribution is higher, the volume anisotropy is lower, resulting in a constant value for the Co layer thickness at which the total anisotropy is zero. We will return to this later on.

Also samples were prepared in which the amount of Co was just enough to form half a monolayer (1 Å). As shown in Fig. 2 the anisotropy falls down because there is no complete interface anymore between Co and Pd. Samples prepared by coevaporation showed a small perpendicular anisotropy, too, probably due to pair ordering during growth. We examined the influence of the local environment of the Co atoms on the anisotropy in these films in a pair interaction model.

A. Pair interaction model

The influence of the surroundings of an atomic magnetic moment on its preferential direction can phenomenologically be described by the interaction energy $w(r, \varphi)$ between all pairs of atoms, depending on the distance r between the two atoms and the angle φ between the direction of the magnetic moment and the line connecting the pair of atoms⁹ (Fig. 3). The magnetic moments are assumed to be oriented parallel by the isotropic exchange interaction. We expand the interaction w as a series and consider the first angle-dependent term $k(r) \cos^2 \varphi$ as the pair energy. Taking only nearest neighbors, the sum over all pairs in the bulk fcc and/or hcp phase is zero, so that to describe the bulk anisotropy more pairs need to be considered. At the interface in the Pd/Co multilayers however, the nearest neighbors of the Co atoms are partially replaced by Pd so that the (large) contributions no longer cancel. For a (111) interface the anisotropy per Co atom becomes

$$w = \sum_i k_i \cos^2 \varphi_i = \frac{3}{4} (k_{Co} - 2k_{Pd}) \sin^2 \Theta = L \sin^2 \Theta, \quad (3)$$

in which i runs over the nearest neighbors; k_{Co} is the interac-

FIG. 3. (a) Definition of the parameters as used in the pair interaction model. The magnetic moment is indicated by the vector m , but does not have to be localized. (b) A schematic, two-dimensional drawing of the relative orientation of the atoms at the interface illustrates the anisotropic bonding of the Co atoms at the interface.

FIG. 4. (a) Temperature dependence of the magnetization of 4.1 Å Co layers with different Pd thicknesses. The average composition of these multilayers is, respectively, 0.45, 0.29, 0.17, and 0.11 Co. The total thickness of each multilayer is 3000 Å. (b) The temperature dependence of the magnetization in PdCo alloys shows a continuous decrease of magnetic order when the Co concentration decreases.

tion between two Co atoms, k_{Pd} between a Co and Pd atom, and Θ is the angle between the magnetic moment and the axis perpendicular to the interface.

Interdiffusion will lower this interface effect, because it increases the randomness of the Co-Pd bonds. Assuming a concentration c of Co atoms in each atomic (111) plane, randomly distributed over the available sites, we get per Co atom an average anisotropy

$$\bar{w} = L \sin^2 \Theta \frac{\sum (2c_j - c_{j-1} - c_{j+1})c_j}{\sum c_j} = L' \sin^2 \Theta, \quad (4)$$

in which the summation runs over all (111) planes. When we consider Co monolayers with perfectly sharp interfaces we have the concentrations $\{c\} = \dots, 0, 1, 0, \dots$ in consecutive layers, resulting in $L' = 2L$. When more interdiffusion or roughness is introduced we get for a nominal monolayer, e.g., $\dots, 0, 0.5, 0.5, 0, \dots$ and $L' = 0.5L$. This illustrates the strong dependence of the interface anisotropy on the mixing at the interface. If $\frac{1}{2}a\sqrt{2}$ is the distance between two nearest-neighbor Co atoms, the area per Co atom at the interface is $S = \frac{1}{4}a^2\sqrt{3}$. Because each layer has two interfaces, the experimental interface anisotropy is now

$$K_s = \frac{L'}{2S} \sum c_j = \frac{1}{2\sqrt{3}} \frac{L'}{a^2} \sum c_j < \frac{1}{4\sqrt{3}} \frac{L'}{a^2}. \quad (5)$$

Taking $a = 3.8$ Å and $K_s = 0.55 \times 10^{-3}$ J/m², this yields $L > 0.2$ meV per Co atom.

When the layers are more interdiffused, the magnetization distribution is more homogeneous, which leads to a lower magnetostatic energy ($\sim M^2$). This causes an increase of K_v and explains the simultaneous change of K_s and K_v as was observed above.

B. Temperature dependence

The temperature dependence of the magnetization of a series of multilayers with a fixed Co thickness (4.1 Å) and varying Pd thickness is given in Fig. 4(a), and compared to equivalent alloys in Fig. 4(b). Despite the fact that the Co layers are only two atoms thick, no linear relation^{10,11} is found. Temperatures above 600 K destroy the layered structure.¹² The long-range interaction across the Pd layers has a range between 13.5 and 27 Å.

III. CONCLUSIONS

The magnetic anisotropy energy in Pd/Co multilayers can be measured most directly by magnetization measurements with the field parallel and perpendicular to the film plane. Phenomenologically the interface contribution to this anisotropy and the effect of interdiffusion at the interface can be described quite well by the anisotropy in Co-Pd bonds. To get a relevant value for the pair interaction an independent measurement of the interdiffusion on atomic scale is necessary, or one should have the ideal case of atomically flat interfaces.

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