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Magnetization curves of Pd/Co multilayers with perpendicular anisotropy

H. J. G. Draaisma^{a)} and W. J. M. de Jonge

Eindhoven University of Technology, Department of Physics, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

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In a ferromagnetic thin film with strong perpendicular anisotropy, saturation may be reached at fields lower than the magnetization. This field is calculated for a multilayer with alternating ferromagnetic and nonmagnetic layers assuming that the stripe domains are oriented only up or down along the anisotropy axis. The results are compared with experimental data on Pd/Co multilayers with ultrathin Co layers. The agreement is very good if we take $\sigma_w = 1 \times 10^{-3}$ J/ m² as the energy of walls between the domains. It is argued also that with other materials, high perpendicular relative remanence may be achieved when multilayers with suitable parameters are used.

I. INTRODUCTION

Multilayer thin films consisting of periodically alternating ferromagnetic and nonmagnetic layers may have interesting new properties. In vapor deposited Pd/Co multilayers, high perpendicular anisotropy has been found when the Co layers are very thin (< 8 Å). This anisotropy can be attributed to the interface between Pd and Co.¹ In rf-sputtered Pd/Co multilayers, a smaller contribution of the interfaces to the total anisotropy was found, probably due to more diffuse interfaces.² Both types of films are put forward to be a candidate for perpendicular recording. The high remanent magnetization may be an important parameter to obtain a high signal-to-noise ratio.³ Furthermore, the coercive field must be suitably matched to the recording heads used.

In this paper we will examine the shape and properties of the perpendicular magnetization curve of multilayers without taking into account any coercivity effects. This means that the domain walls will be assumed to be completely freely mobile. It is further assumed that for low fields the effect of the coercivity can be approximated by shifting the calculated magnetization curve along the field axis. In this way we expect to obtain a high remanence when the perpendicular field in which the magnetization saturates is smaller than the coercive field. The results of the calculations will be compared with experimental data on Pd/Co multilayers, but will in principle be applicable to any multilayer thin film having perpendicular anisotropy in the ferromagnetic layers.

II. PERPENDICULAR MAGNETIZATION CURVES

Consider a multilayer with N bilayers consisting of a ferromagnetic layer of thickness t and a nonmagnetic layer of thickness s. We assume that the perpendicular anisotropy is large enough to orient all (stripe) domains up or down. When this is not the case, closure domains may shortcircuit the magnetic flux. The domain walls are assumed to be infinitely thin and freely mobile, resulting in a periodic domain structure as shown in Fig. 1. In the z direction the domains

are parallel, and in order to obtain a minimum magnetostatic energy the domain walls will be at the same positions in all layers. The magnetization in each domain is the spontaneous magnetization M_s , the length of the domains with the magnetization along the positive z axis is d_1 , and along the negative z axis d_2 , yielding a net magnetization of

$$\tilde{M} = (d_1 - d_2)/(d_1 + d_2)M_s.$$
(1)

The magnetic energy of this domain structure contains three terms: the magnetostatic or demagnetizing energy E_d , originating from the poles at the interfaces between the ferromagnetic and nonmagnetic layers, the wall energy E_w of the domain walls between neighboring domains, and the field energy E_h arising from the interaction of the magnetization with the applied magnetic field. All the energies are calculated per unit volume of the ferromagnetic material and will be normalized to the maximum magnetostatic energy $\frac{1}{2}\mu_0M_s^2$.

In the Appendix we show that the magnetostatic energy can be written as

$$e_{d} = \frac{E_{d}}{\frac{1}{2}\mu_{0}M_{s}^{2}} = m^{2} + \sum_{n=1}^{\infty} \frac{4}{(n\pi)^{3}} \frac{d}{t} \times \sin^{2}[\frac{1}{2}\pi n(m+1)]f_{n}(d), \qquad (2)$$

with

$$f_n(d) = 1 - \exp\left(-2\pi n \frac{t}{d}\right) + \frac{\sinh^2[\pi n(t/d)]}{\sinh^2[\pi n(D/d)]} \times \left\{\frac{1}{N}\left[1 - \exp\left(-2\pi n \frac{ND}{d}\right)\right] - \left[1 - \exp\left(-2\pi n \frac{D}{d}\right)\right]\right\},$$

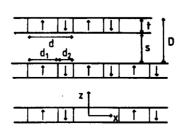


FIG. 1. Model of a domain structure in a multilayer with alternating ferromagnetic and nonmagnetic layers. The domain walls are infinitely thin and freely mobile, resulting in a periodic structure. In the perpendicular direction (z), parallel-oriented domains will give the lowest magnetostatic energy.

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^{a)} In detachment at Philips Research Laboratories, Eindhoven, The Netherlands.

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and with $m = \tilde{M} / M_s$ as relative average magnetization. For $N = 1, f_n(d)$ is identical to the expression derived by Kooy and Enz,⁴ and e_d is independent of s (and D).

Assuming a specific wall energy of σ_w per unit area of a domain wall, we can write

$$e_{w} = \frac{E_{w}}{\frac{1}{2}\mu_{0}M_{s}^{2}} = \frac{2\sigma_{w}}{d(\frac{1}{2}\mu_{0}M_{s}^{2})} = \frac{2\tau}{d},$$
(3)

in which $\tau = \sigma_w / \frac{1}{2} \mu_0 M_s^2$ is a length characteristic for the ferromagnetic material under consideration. τ can assume values ranging from a few nanometers to many micrometers.

Finally, we have '

$$e_{h} = \frac{E_{h}}{\frac{1}{2}\mu_{0}M_{s}^{2}} = \frac{-\mu_{0}HM}{\frac{1}{2}\mu_{0}M_{s}^{2}} = -2hm, \qquad (4)$$

where $h = H/M_s$ is the normalized field applied perpendicular to the film.

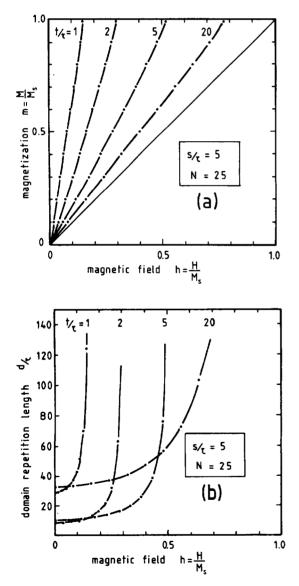


FIG. 2. (a) Perpendicular magnetization curves for multilayers with 25 bilayers (N = 25), a nonmagnetic layer thickness $s/\tau = 5$, and different ferromagnetic layer thicknesses t/τ ; (b) domain repetition length d/τ corresponding to the magnetization curves.

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The total energy $e = e_d + e_w + e_h$ can be minimized with respect to the domain repetition length d and the magnetization m, which is the same as minimizing with respect to the domain lengths d_1 and d_2 :

$$\frac{\partial e}{\partial d} = 0 \rightarrow d = \left[2\tau / \left(\frac{\partial e_d}{\partial d} \right) \right]^{1/2}, \tag{5}$$

$$\frac{\partial e}{\partial m} = 0 \rightarrow h = m + \sum_{n=1}^{\infty} \frac{1}{(n\pi)^2} \frac{d}{t} \sin[\pi n(m+1)] f_n(d) .$$
(6)

The calculation procedure is to choose a magnetization m, calculate d from (5), and find the field at which this magnetization is reached from (6). It is to be noted that all length parameters (t,s,d) can be taken relative to the specific length τ , since only ratios of lengths appear in the formulas. As parameters, we have N, t/τ , and s/τ , which describe the perpendicular magnetization curve completely. The results in this paper were obtained by numeric computation of (5) and (6). Much attention was paid to the slow convergence of the series, especially in the case of large d/τ .

In Fig. 2, a number of curves, giving combinations of h, m, and d/τ , is shown for N = 25, $s/\tau = 5$, and different values of t/τ . The results show a compromise between the opposing tendencies of the individual energy terms: The greater the domain repetition length d/τ , the lower the wall energy will be. The poles of the domains at each side of the nonmagnetic layer also favor a large d/τ , but neighboring poles of reversed domains do the opposite. If there is no magnetic field, the situation is symmetric, $d_1 = d_2$, and d is twice the domain size. The introduction of h along the domain direction increases the size of one kind of domain at the expense of the other, in the first instance without seriously affecting the repetition length d, as can be seen in Fig. 2(b). The minimum of d/τ at h = 0 for a specific value of the ferromagnetic layer thickness t/τ is also observed for single layers.⁵ For N = 1, the curves are independent of s/τ and completely in accordance with those reported for a single layer by Kooy and Enz.⁴

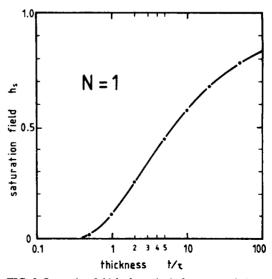


FIG. 3. Saturation field h_s for a single ferromagnetic layer as function of thickness of the layer t/τ . The limiting cases of a multilayer $(s/\tau < 1 \text{ or } s/\tau > 10)$ can be treated as a single film.

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III. SATURATION FIELDS

The concave shape, as in Fig. 2(a), is obtained for any set of parameters. We will now focus our attention on the magnetic field at which saturation is reached. We will consider the field $h_s = H_s/M_s$, at which m = 0.99. Figure 3 shows the increase of h_s with increasing thickness t/τ for a single ferromagnetic layer. In Fig. 4 the dependence of h_s on the thickness of the nonmagnetic layers in a multilayer, s/τ , is shown for different N and t/τ . In the limit of large s/τ and arbitrary N we find the same value for h_s as for N = 1, because the layers are magnetostatically decoupled. For very small values of s/τ the multilayer behaves magnetically as a single ferromagnetic layer of thickness $N(t/\tau)$. The transition between these two limiting cases takes place for $s/\tau = 1-10$.

IV. COMPARISON TO EXPERIMENTAL DATA

The results derived above can be compared with experimental data which have been obtained on vapor-deposited Pd/Co multilayers. Only in the cases of 2-Å and 4-Å Co is

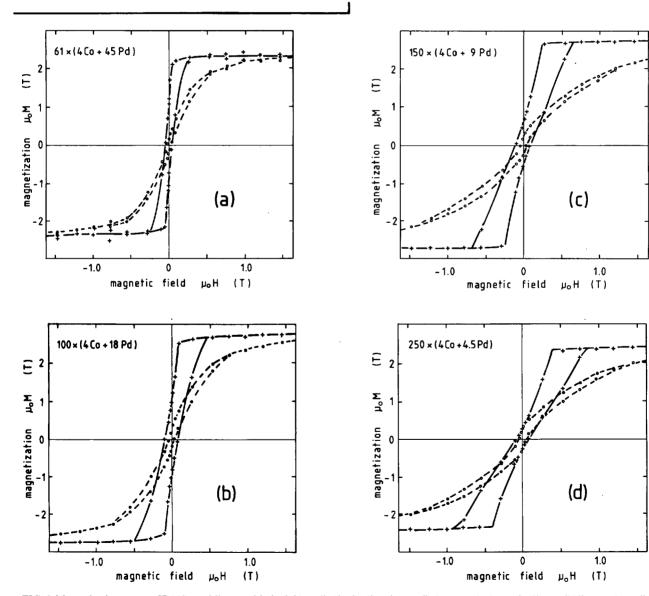


FIG. 5. Magnetization curves of Pd/Co multilayers with the field applied in the direction applied perpendicular to the film (solid line) and parallel to the film (dashed line). The films have 4-Å Co layers and different Pd thicknesses: (a) 45-Å Pd, (b) 18-Å Pd, (c) 9-Å Pd, and (d) 4.5-Å Pd.

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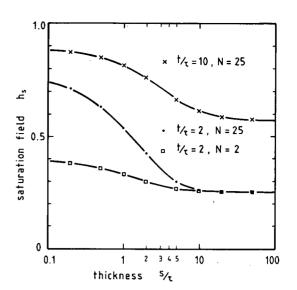


FIG. 4. Saturation field h_s of a multilayer as a function of the thickness of the nonmagnetic layers s/τ for different values of the ferromagnetic layer thickness t/τ and the number of bilayers N.

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TABLE I. Comparison of the saturation fields of vapor-deposited Pd/Co multilayers h_s (exp), as estimated from the magnetization curves, with the calculated fields h_s (calc). A good agreement is found for $\mu_0 M_s = 1.76$ T, as for bulk Co, and $\tau = 8$ Å. The resulting domain-wall energy is $\sigma_w = 1 \times 10^{-3}$ J/m².

t _{Co} (Å)	t _{Pd} (Å)	N	$\mu_0 H_s(T)$	h _s (exp)	t/τ	s/T	$h_s(\text{calc})$
4	4.5	250	0.69	0.39	0.5	0.56	0.404
4	9	150	0.45	0.26	0.5	1.13	0.254
4	18	100	0.27	0.15	0.5	2.25	0.147
4	45	61	0.12	0.07	0.5	5.63	0.065
2	4.5	300	0.38	0.22	0.25	0.56	0.254
2	6.7	250	0.36	0.20	0.25	0.84	0.186
2	9	200	0.25	0.14	0.25	1.13	0.144
2	11.2	200	0.23	0.13	0.25	1.4	0.121
2	13.5	150	0.20	0.11	0.25	1.69	0.100
2	18	100	0.15	0.09	0.25	2.25	0.079

the anisotropy in the Co layers large enough to ensure that only up and down domains will exist. In Fig. 5 we show a number of magnetization curves for multilayers with 4-Å Co and different Pd thicknesses. The determination of the saturation field is somewhat complicated by the observed hysteresis, but the decrease of H_s with increasing Pd thickness is obvious. H_s is estimated by drawing a line through the origin parallel to both branches of the perpendicular magnetization curve; the results are given in Table I. Using for the magnetization of the Co layers the same value as for bulk Co, as seems to be justified by previous measurements,¹ we obtain a good agreement for $\tau = 8$ Å, which yields for the energy of a domain wall in these layers $\sigma_w = 1 \times 10^{-3} \text{ J/m}^2$. Also, for multilayers with 2-Å Co layers the comparison, using the same parameters, is very satisfactory. It is the low value of H_s combined with a large coercive field which results in a high remanence (0.95-0.99) for these multilayers.³

The direct comparison of a calculated curve with experimental data is complicated by the hysteresis, but they both show the same concave shape. The quantitative agreement between theoretical results and experimental data for this system is somewhat surprising in view of the rather crude approximations we have used. In fact, the actual situation in the Pd/Co multilayers is much more complicated. To explain the observed magnetization, polarization of some of the Pd atoms was introduced,³ whereas in our model Pd is treated as a vacuum. Furthermore, light scattering experiments on Fe-Pd-Fe films indicate an exchange coupling over Pd layer thicknesses up to 30 Å.⁶ Although this additional coupling is not incompatible with our model (since the domains at both sides of a Pd layer are parallel), it might influence the value of the domain-wall energy and thus the value of τ .

V. CONCLUSION

The effect of the Pd thickness on the magnetization of a Pd/Co multilayer with perpendicular anisotropy can be un-

derstood both qualitatively and quantitatively by the magnetostatic interaction between the perpendicular domains. By choosing a suitable combination of parameters, the saturation field in such multilayers can be reduced, resulting in a high relative remanent magnetization when the coercive field is sufficiently large.

It may also be fruitful to make multilayers in the case of other perpendicular recording materials, such as CoCr. The relative remanent magnetization of a single-layer CoCr of several hundreds of nanometers is approximately 0.05–0.1.⁷ To decrease H_s the layer thickness should be reduced, and to retain the total magnetic moment, one might consider a multilayer containing such thin ferromagnetic layers. Because the perpendicular anisotropy in CoCr is of crystalline origin and for this material $\tau \approx 100-200$ Å (10–20 nm), the CoCr layers in the multilayer do not have to be as extremely thin as in the Pd/Co case, in which the perpendicular anisotropy stems from the interfaces between Pd and Co.

From this model, predictions about the domain sizes can be made in partially magnetized as well as in demagnetized states. It would be very interesting to compare these with observations from direct domain imaging techniques.

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APPENDIX

The magnetostatic energy of domains oriented perpendicular to a thin film or plate has been treated by several authors: Kittel⁸ considered a homogeneous ferromagnetic film in the limit of noninteracting sides and in the demagnetized state in which up and down domains are of equal size. Málek and Kambersky⁵ included the interaction between the sides of the plate. Kooy and Enz⁴ extended the calculation to magnetized states. Very recently, Suna⁹ considered the problem of magnetostatic interactions in a multilayer, but only in the demagnetized state. For clarity and to avoid notational problems, we will give a survey of the entire calculation.

To calculate the magnetostatic energy of a domain configuration as shown in Fig. 1, we first need the potential $\phi(H = -\nabla\phi)$ of a single layer, which satisfies the Laplace equation

$$7^2\phi = 0, \tag{A1}$$

with the proper boundary conditions. We distinguish between the regions outside the layer $[\phi_0(x,z), z > \frac{1}{2}t]$ and inside the layer $[\phi_i(x,z), -\frac{1}{2}t \le z \le \frac{1}{2}t]$. By antisymmetry in z, we have for $z < -\frac{1}{2}t$, $\phi(x,z) = -\phi_0(x, -z)$. According to this model, we write the magnetization as

$$M(x,z) = M_s, \quad -\frac{1}{2}d_1 \leqslant x < \frac{1}{2}d_1 \qquad \Lambda \qquad -\frac{1}{2}t \leqslant z < \frac{1}{2}t, \\ -M_s, \quad \frac{1}{2}d_1 \leqslant x < \frac{1}{2}d_1 + d_2 \qquad \Lambda \qquad -\frac{1}{2}t \leqslant z < \frac{1}{2}t, \\ 0, \qquad z < -\frac{1}{2}t \qquad V \qquad z > \frac{1}{2}t, \end{cases}$$

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periodic in x over a distance $d = d_1 + d_2$: M(x + d,z) = M(x,z). Written as a Fourier series, this becomes $(-\frac{1}{2}t \le z \le \frac{1}{2}t)$

$$M(x,z) = \widetilde{M} + \sum_{n=1}^{\infty} \frac{4M_s}{\pi n} \sin[\frac{1}{2}\pi n(m+1)] \cos\left(2\pi n\frac{x}{d}\right),$$
(A3)

in which $\widetilde{M} = (d_1 - d_2/d)M_s$ is the average magnetization and $m = \widetilde{M}/M_s$. As boundary conditions, we now have

$$-\frac{\partial\phi_0}{\partial z}(x, \frac{1}{2}t) = -\frac{\partial\phi_i}{\partial z}(x, \frac{1}{2}t) + M(x, \frac{1}{2}t),$$

$$\phi_0(x, \frac{1}{2}t) = \phi_i(x, \frac{1}{2}t).$$
(A4)

A general solution of Eq. (A1), taking into account the shape of the (x,z) region to which it should apply, can be written as

$$\phi(x,z) = A_0 + B_0 z + \sum_{k=1}^{\infty} \left[A_k \cos\left(2\pi k \frac{x}{d}\right) \exp\left(-2\pi k \frac{z}{d}\right) + B_k \cos\left(2\pi k \frac{x}{d}\right) \exp\left(2\pi k \frac{z}{d}\right) \right].$$
(A5)

If we substitute (A5) into (A4) we get

$$\phi_0(x,z) = \frac{1}{2}\widetilde{M}t + \sum_{k=1}^{\infty} \frac{2M_s d}{(\pi k)^2} \sin\left[\frac{1}{2}\pi k(m+1)\right] \\ \times \sinh\left(\pi k \frac{t}{d}\right) \cos\left(2\pi k \frac{x}{d}\right) \exp\left(-2\pi k \frac{z}{d}\right),$$

$$\phi_i(x,z) = \widetilde{M}z + \sum_{k=1}^{\infty} \frac{2M_s d}{(\pi k)^2} \sin\left[\frac{1}{2}\pi k(m+1)\right] \\ \times \exp\left(-\pi k \frac{t}{d}\right) \cos\left(2\pi k \frac{x}{d}\right) \sinh\left(2\pi k \frac{z}{d}\right).$$
(A6)

Now, the multilayer problem with parameters as in Fig. 1 can be solved. By superposition, the total potential $\phi_p(x,z)$ in layer *p*, originating from all the *N* layers, with z = 0 in the center of layer *p* and $\phi_p(x,0) = 0$ is

$$\phi_{p}(x,z) = \sum_{j=1}^{p-1} \{\phi_{0}[x,z+(p-j)D] \}$$
$$-\phi_{0}[x,(p-j)D] \} + \phi_{i}(x,z)$$
$$-\sum_{j=p+1}^{N} \{\phi_{0}[x,(j-p)D-z] \}$$
$$-\phi_{0}[x,(j-p)D] \}, \qquad (A7)$$

with the minus sign resulting from the antisymmetry. The corresponding energy density involved for layer p is

$$E_{p} = \frac{\mu_{0}}{2dt} \int_{0}^{d} dx \, M(x, \frac{1}{2}t) \left[\phi_{p}(x, \frac{1}{2}t) - \phi_{p}(x, -\frac{1}{2}t) \right].$$
(A8)

Inserting (A6) and (A7) and using the Fourier series (A3), the only terms that remain after the integration over x are those with n = k:

$$E_{p} = \frac{1}{2}\mu_{0} \left\{ \widetilde{M}^{2} + \sum_{n=1}^{\infty} \frac{4M_{s}^{2}}{(\pi n)^{3}} \frac{d}{t} \sin^{2} \left[\frac{1}{2}\pi n(m+1) \right] \left[1 - \exp\left(-2\pi n \frac{t}{d} \right) - 2\sinh^{2}(\pi n \frac{t}{d}) \left(\frac{\exp\left[-2\pi n(p-1)D/d \right] + \exp\left[-2\pi n(N-p)D/d \right] - 2}{1 - \exp(2\pi nD/d)} \right) \right] \right\}.$$
(A9)

Normalizing the energy to $\frac{1}{2}\mu_0 M_s^2$, the maximum magnetostatic energy, and averaging over all the layers, we have

$$e_d = (1/N) \left(\sum_{p=1}^N E_p / \frac{1}{2} \mu_0 M_s^2 \right),$$
 (A10)

resulting in expression (2) given in the text.

1

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