Oscillatory interlayer exchange coupling in Co/Ru multilayers and bilayers

P.J.H. Bloemen
Department of Physics, Eindhoven University of Technology (EUT), 5600 MB Eindhoven, The Netherlands

H.W. van Kesteren
Philips Research Laboratories, Professor Holstlaan 4, 5656 AA Eindhoven, The Netherlands

H.J.M. Swagten and W.J.M. de Jonge
Department of Physics, Eindhoven University of Technology (EUT), 5600 MB Eindhoven, The Netherlands
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The interlayer exchange coupling in several Co/Ru multilayers and bilayers prepared by sputtering has been investigated from magnetization and ferromagnetic resonance experiments. The hysteresis loops have been discussed in detail, particularly regarding their dependence on the number of layers and the effect of an easy axis direction perpendicular to the film plane. Ferromagnetic as well as antiferromagnetic coupling strengths have been determined in an extended range of Ru thicknesses up to 44 Å. The analysis yields an oscillatory magnetic coupling as a function of the Ru thickness having a period of approximately 12 Å. The peak-to-peak-distance has been found to be smaller at thinner Ru layers indicating a preasymptotic or multiperiodic behavior in the coupling. The envelope function of the coupling was found to decrease, initially as t_{Ru}^{-2} but considerably faster above a Ru thickness of 20 Å. The latter is discussed in terms of mean-free-path effects and of Fermi surface smearing effects both of which are active as a result of structural defects and of the finite measuring temperatures.

I. INTRODUCTION

The interesting and sometimes spectacular novel phenomena displayed by artificially layered magnetic materials have drawn considerable attention in recent years. In particular the large perpendicular magnetic anisotropy,[1] the (giant) magnetoresistance,[2–4] and the (generally) oscillating interlayer interaction[5] have been the subject of fundamental as well as technological interest.

Usually the antiferromagnetic (AF) interlayer coupling is studied using magnetic layers having an in-plane preferential orientation. The field dependence of the magnetization then shows a linear curve saturating at a field which is directly proportional to the strength of the antiferromagnetic coupling. Less common are studies in which the layers exhibit a strong uniaxial magnetic anisotropy. The latter can be realized, for example, in layers with an easy axis perpendicular to the plane. The presence of such perpendicular anisotropy might prove convenient to remove certain complicating effects to which in-plane magnetic anisotropy systems are sensitive. For example, perpendicular alignment of the magnetizations due to biquadratic coupling effects (see, for example, Ref. 6) are not likely to occur in the case of strong uniaxial anisotropy since this will be energetically unfavorable. Also, contributions of locally ferromagnetically coupled regions in the sample (e.g., due to spacer layer fluctuations) are expected to have considerably less influence on the magnetization curves of AF-coupled layers with uniaxial anisotropy.[7] For samples with an in-plane preferred direction for the magnetization such regions immediately give rise to a nonzero remanence, thereby obscuring the AF coupling effect. Systems with perpendicular magnetic anisotropy are therefore expected to reveal AF coupling effects more clearly.

In this paper results are presented of investigations on the magnetic coupling between Co layers across Ru spacer layers with emphasis on those having a perpendicular magnetic anisotropy. Apart from magneto-optical Kerr effect (MOKE) and vibrating sample magnetometer (VSM) measurements to determine the AF coupling, ferromagnetic resonance (FMR) experiments have been performed to determine the ferromagnetic coupling. MOKE measurements have been carried out on the samples with a perpendicular preferential orientation, whereas the VSM and FMR experiments have been performed for in-plane magnetized Co/Ru/Co trilayers. The field-induced magnetic phase transitions, which include information about the interlayer interaction, are critically evaluated, specifically with regard to their dependence on the number of ferromagnetic layers involved. Moreover, a comparison is made between the loops of the present *sputtered* samples and the loops of similar samples grown by *evaporation.* Marked differences are observed, demonstrating that the preparation method is extremely important for the magnetic behavior.

The paper is organized as follows. In Sec. II the experimental details regarding the samples are contained. Section III briefly recalls the theoretical thermodynamic phase transitions expected for AF-coupled magnetic layers with uniaxial magnetic anisotropy. This is followed by Sec. IV which considers the effects of the number of

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TABLE I. Detailed composition of the samples which have been investigated. "Ox Si" is short for oxidized silicon. The numbers in the last column indicate the range of Ru thicknesses for which the samples have been grown.

Series	Substrate	Base layers + active layers + cap layers	x (Å)
A	Ox Si	200 Å Ru + 9 Å Co/x Å Ru/9 Å Co + 30 Å Ru	8,16–48
B	Ox Si	200 Å Ru + 6×(9 Å Co/18 Å Ru) + 12 Å Ru	
	Ox Si	200 Å Ru + 10×(9 Å Co/18 Å Ru) + 12 Å Ru	
	glass, Ox Si	500 Å Pd/200 Å Ru + 10×(11 Å Co/x Å Ru)	3–31
C	glass	200 Å Pd + 30 Å Co/x Å Ru/70 Å Co + 50 Å Pd	4–51

repetitions on MOKE loops for multilayers with a varying number of bilayers. Section V discusses the experiments aimed at determination of the magnetic interlayer coupling. This section is divided into three parts containing (A) the MOKE experiments on the samples with a perpendicular preferential orientation, (B) the VSM and FMR experiments on the in-plane easy Co/Ru/Co trilayers, and (C) a discussion of the results. The paper is summarized briefly in Sec. VI.

II. EXPERIMENTAL DETAILS

The compositions of the series of samples which have been investigated are listed in Table I. In the following the series will be referred to as series A, B, and C, as indicated in the first column of the table. All samples were prepared at room temperature by magnetron sputtering (Ar) at a background pressure of 4×10^{-7} Torr with sputtering rates of typically 1–2 Å/s. The substrates were given a 30 min glow-discharge treatment prior to the base layer deposition. Contrary to all other layers, the Pd base layers were prepared by rf sputtering. The layer thicknesses were calibrated from low angle x-ray diffraction experiments on reference samples of approximately 500 Å thick Co, Pd, and Ru layers which were prepared preceding and following the preparation of the actual samples. The layer thicknesses of these calibration samples reproduced within 5%. In the case of multilayers the actual modulation periods have been determined from the positions of the low angle multilayer reflections; see Fig. 1(a) for a typical example. From the positions of the high angle reflections [e.g., Fig. 1(b)], a fcc (111) or hcp (00.2) texture has been concluded. However, the texture was not very well defined. The rocking curves of both the Pd and Ru base layers and of the multilayer appeared rather broad (13° full width at half maximum).

Magnetic characterization has been performed at room temperature using the magneto-optical Kerr effect (MOKE) at a wavelength of 633 nm, ferromagnetic resonance (FMR) at 34 GHz, and vibrating sample magnetometry (VSM).

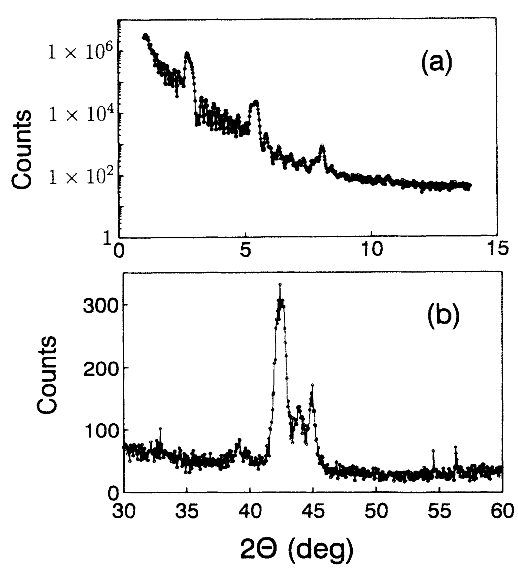

FIG. 1. (a) Low and (b) high angle x-ray diffraction pattern (λ = 1.5418 Å) of a 200 Å Ru + 10×(9 Å Co/24 Å Ru) + 12 Å Ru multilayer grown by sputtering on an oxidized Si substrate.

III. THEORETICAL MAGNETIZATION CURVES

Many of the samples listed in Table I exhibit an easy axis perpendicular to the film plane. Since such an easy axis has a pronounced effect on the magnetization curves, this section recalls the most important results of previous calculations[8,9] for the magnetization curves in these cases.

The starting point is the energy expression (J/m^2) for a bilayer:

$$E = -\mu_0 M_s H t[\cos\theta_1 + \cos\theta_2] - Kt[\cos^2\theta_1 + \cos^2\theta_2] -2J\cos(\theta_1 - \theta_2). \quad (1)$$

Here, the bilayer consists of two identical magnetic layers of thickness t, which are coupled antiferromagnetically with a strength J ($J < 0$ J/m^2), having uniaxial magnetic anisotropy K and saturation magnetization M_s. The angles of the magnetizations with respect to the film normal are denoted by θ_i ($i = 1,2$). The applied field H is directed along the easy axis, in our case the film normal ($K > 0$). The case of more than two magnetic layers will be discussed later on.

Absolute minimum energy calculations[8,9] giving the thermodynamically stable states and corresponding transitions result in magnetization curves shown in Fig. 2. Here the shape of the magnetization curve depends on the ratio between the strength of the magnetic anisotropy and the strength of the antiferromagnetic coupling. For $K > -J/t$ and $K < -J/t$ the respective curves shown in Fig. 2(a) and 2(b) apply. The curve in Fig. 2(c) is a special case of the one shown in Fig. 2(b). The charac-

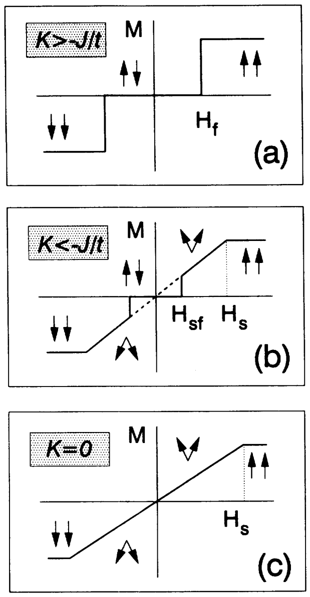

FIG. 2. Theoretical magnetization curves for two identical AF-coupled ferromagnetic layers having a uniaxial magnetic anisotropy which is (a) larger than the coupling strength and (b) smaller than coupling strength. The arrows schematically indicate the orientations of the magnetizations relative to the vertical easy axis, along which the field is applied. The situation of zero anisotropy (c) is a special case of (b).

teristic fields H_f, H_{sf}, and H_s occurring in Figs. 2(a), 2(b), and 2(c), are given by

$$H_f = -2J/t\mu_0 M_s \qquad (K > -J/t), \tag{2}$$

$$H_s = -2(K + 2J/t)/\mu_0 M_s \qquad (K < -J/t), \tag{3}$$

$$H_{sf} = 2\sqrt{-K(K + 2J/t)}/\mu_0 M_s \qquad (K < -J/t), \tag{4}$$

$$H_s = -4J/t\mu_0 M_s \qquad (K = 0). \tag{5}$$

As was mentioned, these formulas were derived from absolute minimum energy calculations; no hysteresis occurred. However, if one allows for coherent rotation of the magnetic moments only and takes the magnetic layers always to be in a single domain state, i.e., excluding the mechanisms which usually drive the system to the state of absolute minimum energy, such as domain nucleation and domain wall propagation, the above results will be modified. The curves neccessarily show hysteresis due to the existence of energy barriers resulting from the magnetic anisotropy. The spin-flip and spin-flop fields are different when approached from the low or high field side. In Refs. 8 and 9 the resulting fields have been derived by evaluation of the usual stability condition corresponding to the appearence of a path on the energy surface $E(\theta_1, \theta_2)$ which permits the system to slide from its current stable state into the most accessible *local* minimum. Since it will appear that these results are not applicable in our case because the actual hysteresis effects are much smaller than calculated, these results will not be recalled here.

The case of *multilayers* is expected to be more complicated due to boundary effects. Even in the limit of an infinite number of layers the relevant transitions cannot be obtained by simple substitution of $2J$ for J, in the bilayer expressions. This approximation, which one often applies to multilayers with a large number of repetitions, is allowed only for zero magnetic anisotropy.

Consider a multilayer consisting of an even number of AF-coupled ferromagnetic layers and assume the large K limit. Contrary to the bilayer case, we expect the magnetization curve to exhibit *two* transitions. At low fields first one of the outer layers, viz., the one which is oriented antiparallel to the applied field, will reverse its direction. This is because this layer has one neighboring magnetic layer only and therefore experiences one coupling only. At the second transition the inner layers which are still antiparallel to the field will reverse their direction. The fields pertaining to these spin flips are easily found. The first one is obviously again given by Eq. (2), whereas the second one occurs at twice this field ($-4J/t\mu_0 M_s$). Thus, the fields are *independent* of the number of ferromagnetic layers — this in marked contrast to the isotropic case ($K = 0$) where the saturation field does depend on the number of magnetic layers, n, and was found experimentally to vary approximately as

$$H_s = -8J\left(1 - \frac{1}{n}\right)/t\mu_0 M_s \qquad (K = 0); \tag{6}$$

see Ref. 10. However, this equation is not exact. In fact, as shown in Ref. 8, a closed formula cannot be derived. This is also the case for medium K (spin-*flop* transitions). Numerical calculations are required since the boundary effects cause the canting angles to vary across the layer stack. We have performed such calculations and it appeared that the lowest spin-flop field for the ten repetitions case, i.e., the case applying to the present multilayer samples, deviated only slightly (less than 10% depending on the $-J/Kt$ ratio) from the bilayer value.

IV. MOKE LOOPS FOR VARIOUS NUMBERS OF REPETITIONS

The experiments described in the present paper have been performed on bilayers (sandwiches) as well as on multilayers. Since the identification and characterization of the field-induced phase transitions discussed in the previous section are of crucial importance for the determination of the interlayer exchange, the magnetization process of multilayers with a small number of repetitions has been investigated in somewhat more detail. The full results of this study will be published elsewhere, while here results which are of particular relevance for the present study of the interlayer exchange will be presented.

In Fig. 3, examples of experimental hysteresis loops are shown for 9 Å Co/18 Å Ru multilayers containing two, six, and ten Co layers, measured in perpendicular applied fields. The existence of an antiparallel alignment of the

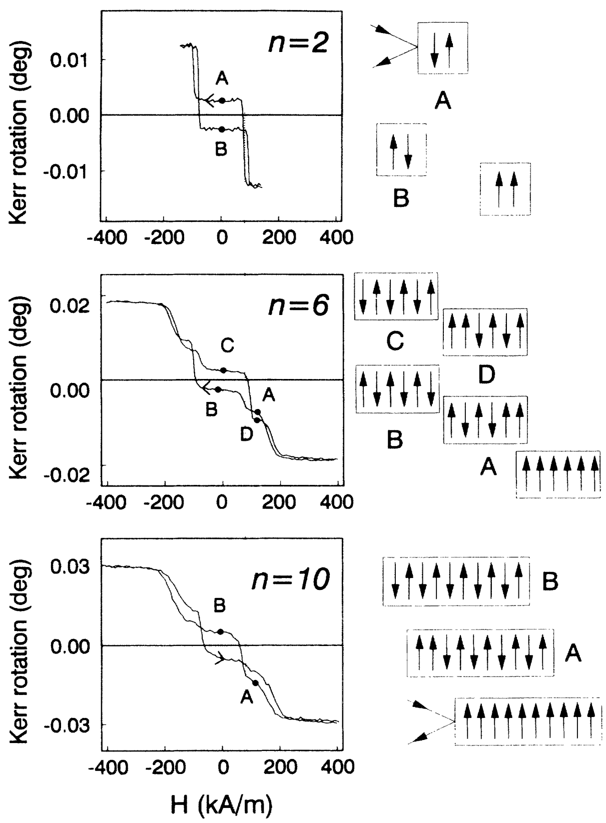

FIG. 3. Hysteresis loops of 200 Å Ru + $n\times$(9 Å Co/24 Å Ru) + 12 Å Ru multilayers with several number of repetitions n, measured with MOKE in perpendicular applied fields. The arrows schematically indicate the orientations of the magnetizations (in reality perpendicular to the film plane) at progressive stages (A,B,C,D) of the magnetization process.

magnetizations along the film normal at low fields is clear from the small Kerr signal. The fact that the Kerr signal is not zero in this state is a result of the finite penetration depth of the light.

To clarify the form of the loops in more detail, the spin configurations corresponding to specific Kerr levels are also shown in the figure. Here, the convention is used that the light "illuminates the spin configuration" from the left. The left arrows within the boxes therefore correspond to Co layer magnetizations at the top of the sample (the surface) and the right ones to the Co layers at the bottom (substrate side).

For the bilayer ($n = 2$) case, reduction of the field from a large value to zero results in a transition from saturation to an antiparallel state. The shape of the loop closely resembles the theoretical loop of Fig. 2(a). Obviously, the interlayer exchange coupling energy is smaller than the magnetic anisotropy energy at this Ru thickness. The change in polarity of the Kerr signal which accompanies the saturated-antiparallel transition indicates that it must have been the top Co layer that reversed its magnetization.

For the $n = 6$ multilayer case, two transitions occur. Upon reducing the field from a large positive value, the first two inner Co layers reverse their magnetization (they are coupled twice). The resulting state is indicated by configuration A. Upon further reduction of the field, the *bottom* outer layer flips which is consistent with the fact that this flip is accompanied by a relatively small though clear step in the Kerr signal. The antiparallel configuration B is the one pertaining to the resulting state. Applying a negative field until saturation and reducing it back to zero yield the reversed antiparallel state C. Increasing the field in the positive direction results now in the reversal of neccessarily the *top* outer layer and is thus accompanied by a *larger* step. The Kerr value of the resulting state D is, as expected, slightly more negative than for state A and thus leads to a crossing in the Kerr loop.

The shape of the loop of the $n = 10$ multilayer, i.e., the existence of a relatively sharp step at low fields in the descending part of the loop (from saturation to zero field, A→B) and its apparent absence in the ascending part, can now be understood. The asymmetry in the low field step size which appeared between the magnetizing and demagnetizing process in the $n = 6$ case is obviously significantly larger for $n = 10$. Apparently it has become so large that a reversal of the bottom outer layer hardly results in any change of the Kerr signal.

Comparing the flip fields (of the outer layer pair) measured with decreasing field for $n = 2$, 6, and 10 (the dotted vertical line), one notes that these agree rather well. This shows that this flip field does not in fact depend on the number of repetitions, as predicted for the large K limit. However, the second transition, predicted at twice this field, is for both the $n = 6$ and the $n = 10$ case, much less sharp than the first one. This phenomenon is related to the fact that this transition involves the reversal of several magnetic layers with possibly slightly differing magnetic properties and/or coupling strength, in contrast to the low field transition involving only one layer.[11] This fact makes this second transition field less suitable to determine the AF coupling strength. However, the *independence* of the lowest flip field on the number of repetitions permits multilayers, which *a priori* seem too complicated due to assumed boundary effects to be used for investigation of the interlayer coupling.

V. INTERLAYER COUPLING

From the theoretical considerations mentioned in Sec. III, it is clear that the magnetic anisotropy energy and the saturation magnetization are important parameters for the detailed analysis of the magnetization curves and the quantitative determination of the interlayer coupling.

As for the magnetization, VSM measurements on series B and the $n = 10$ multilayer of series A have made it clear that the average saturation magnetization per unit Co volume at room temperature was reduced by a factor of 2.8 with respect to the bulk value. A comparable reduction (factor 2.1) for approximately the same Co layer thickness (10 Å) was found by Sakurai *et al.*[12] The most significant part of these reductions is probably due to interdiffusion at the Co/Ru interface, combined with a hybridization between the Co 3*d* and the Ru 4*d* bands. The Curie temperatures as observed by Sakurai *et al.* are too high to account for the reductions: $T_c \approx$ 550 K for 10×(10 Å Co + 20 Å Ru).

The magnetic anisotropy has been determined using MOKE on samples exhibiting no AF coupling. For the detailed measuring procedure the reader is referred to Purcell *et al.*[13] For series A as well as series B a perpendicular anisotropy has been observed, as expected for these ultrathin Co layers because of the contribution of the Co/Ru interface anisotropy.[12,14,15] The anisotropy was determined to be $K = 150 \pm 20$ kJ/m^3. This value is comparable to the 70 kJ/m^3 observed for the molecular-beam-epitaxy- (MBE-) grown 31×(10 Å Co/32 Å Ru) multilayer by Dinia *et al.*,[14] the 300 kJ/m^3 found by Sakurai *et al.* for a vapor deposited 10×(10 Å Co/20 Å Ru) multilayer,[12] and the 120 kJ/m^3 found by the authors previously for 10×(10 Å Co/*x* Å Ru) multilayers which were also grown by evaporation.[15]

We conclude that for Co layers in the order of 10 Å, as used in part of the present experiments, a change from spin-flip behavior to spin-flop behavior can be expected for $-J = Kt \approx 0.15$ mJ/m^2.

A. MOKE hysteresis loops

To investigate the interlayer coupling, hysteresis loops of the samples in Table I have been measured. In Fig. 4 typical magneto-optically measured loops are shown for series A, i.e., samples with 9 Å Co/*x* Å Ru/9 Å Co trilayers. The applied field was oriented perpendicular to the film plane. The square loops (with 100% remanence) observed for Ru thicknesses of 22, 36, and 46 Å again confirm the perpendicular easy axis. The loops are characteristic for ferromagnetically (or weak AF) coupled layers. However, the loops observed for Ru thicknesses of 20, 32, and 44 Å display very clearly the existence of an antiferromagnetic coupling. Typical MOKE loops obtained for the 10×(11 Å Co/*x* Å Ru) multilayers are shown in Fig. 5. It is clear that, in both the bilayer and the multilayer cases, the behavior as a function of the Ru thickness is consistent with an oscillatory exchange coupling. Deduction of the AF coupling strengths from the transition fields is not self-evident. This is due to the hysteresis in the transition fields. Application of the formulas derived by Dieny *et al.*[8] from stability conditions assuming uniform coherent rotation of the magnetizations will yield erroneous results since the *measured* hysteresis is much smaller than the calculated one and equals approximately the single layer hysteresis (see Fig. 4). This is completely analogous to the general observation that coercive fields of single ferromagnetic layers are always much smaller than the corresponding anisotropy fields (the Brown paradox[16]). One is therefore often led to average the hysteresis and apply to the result the formulas derived from absolute minimum energy calculations. It is not clear that this procedure always yields the correct results. In general, the average of the spin-flip fields derived from stability do *not* correspond to the spin-flip fields based on global minimum energy solutions. How-

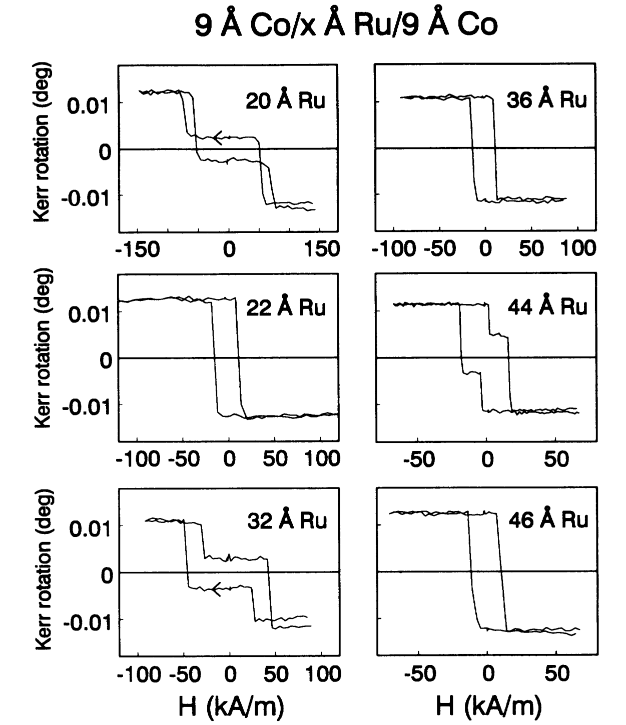

FIG. 4. Hysteresis loops measured with MOKE on sputtered samples of the type 200 Å Ru + 9 Å Co/*x* Å Ru/9 Å Co + 30 Å Ru with Ru thicknesses *x* as indicated. The field is applied along the film normal.

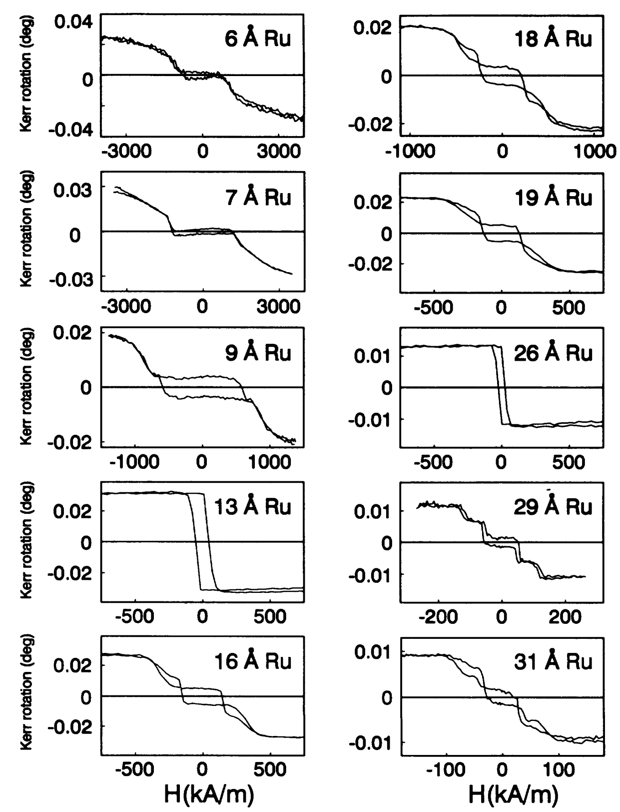

FIG. 5. Magneto-optically measured hysteresis loops of 500 Å Pd + 200 Å Ru + 10×(11 Å Co/*x* Å Ru) multilayers with Ru thicknesses as indicated. The field is applied perpendicular to the film plane.

ever, since calculations of real hysteresis quantities will be complex because of their dependence on the microstructure of the sample and of questionable validity due to the often unknown (magnetic) properties of defects or deviations of perfect structure in general, averaging appears, so far, the most (if not the only) useful procedure. Also in the present case, the average transition fields have been used.

To obtain values for the interlayer interaction J from the data in Fig. 4 and Fig. 5, the two regimes $-J/t < K$ and $-J/t > K$ had to be identified. The first AF region (around 8 Å Ru) corresponds to the regime $-J/t > K$. This can be seen from the shape of the hysteresis loops in this peak; see Fig. 5. The loops at 6 and 7 Å Ru clearly give no indication of a second level. Instead, the relatively small jump (or fast increase) is followed by a steady increase of the Kerr signal — a behavior which closely resembles the theoretical loop in Fig. 2(b). Unfortunately, our available fields were not large enough to obtain saturation for these strongly AF-coupled samples. The coupling values for this peak have therefore been calculated using Eq. (4). This is, as we have mentioned, a good approximation. It should be noted here that, although no loops are shown for Ru thicknesses between 3 and 6 Å, our MOKE measurements in that interval indicated that the coupling is very strongly AF there. The fields, however, were too poorly defined to determine the coupling values reliably. We will come back to this point later on. For larger Ru thicknesses the coupling has reduced considerably and the regime $-J/t < K$ is reached. The coupling strengths for these peaks have been calculated, for both the multilayers and the bilayers, from the first flip field [Eq. (2)]. (The resulting values have been plotted as a function of the Ru thickness in Fig. 9. This figure also contains other data points and will be discussed later on.)

B. FMR and VSM experiments

So far, the information was restricted to AF coupling only. The square hysteresis loops observed in Fig. 4 and Fig. 5 provide no information on the ferromagnetic coupling behavior at the corresponding Ru thickness intervals. To measure the ferromagnetic coupling ferromagnetic resonance (FMR) has been applied. This requires the layers to have *different* magnetic properties since otherwise the intensity of the so-called optical mode is zero and the coupling cannot be determined.[17,18] For this reason the samples of series A and B could not be used for the FMR experiments and a specially designed series of samples has been grown, viz., series C.

The samples consist of two Co layers with thicknesses of 30 and 70 Å, respectively, which are separated by a Ru spacer of varying thickness. Due to the nonzero magnetic interface anisotropy for Co/Ru, the different Co thicknesses correspond to different effective magnetic anisotropies.

In Fig. 6(a) the resonance fields are plotted versus the Ru thickness. The lower and upper dotted horizontal lines indicate the average resonance fields of the decoupled 70 Å and 30 Å Co layers, respectively. The data

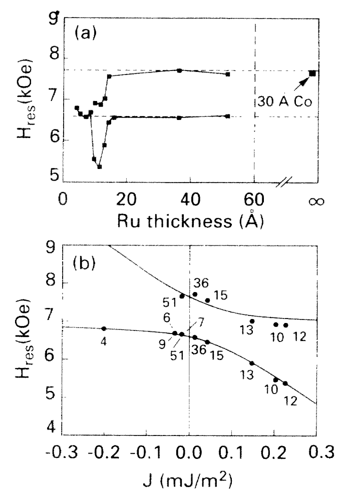

FIG. 6. (a) Resonance fields versus the Ru thickness measured with FMR at 34 GHz with in-plane applied fields on 200 Å Pd + 30 Å Co/x Å Ru/70 Å Co + 30 Å Pd samples. The solid line is a guide to the eye. (b) Fit of the experimental resonance fields (solid circles) to the calculated dependence of the resonance fields on the interlayer exchange coupling (solid lines). The numbers indicated are the Ru thicknesses.

point plotted in the right part of Fig. 6(a) pertained to the sample containing the 30 Å Co layer only: 200 Å Pd/30 Å Co/50 Å Pd. From the figure one can immediately conclude the existence of a maximum in the ferromagnetic coupling at about 12 Å Ru (which is in agreement with the observation of simple square hysteresis loops in the MOKE experiments). At smaller Ru thicknesses (below 10 Å), only one resonance was observed which has shifted to *higher* fields corresponding to AF coupling. This is consistent with the observation of AF coupling from the Kerr loops of the previous series of samples in the same Ru thickness interval. The absence of the second resonance is probably due to the relatively strong AF coupling in this thickness interval, since the optical mode intensity decreases rapidly with increasing coupling strength.[17,18] With these FMR data, the true oscillatory nature of the coupling between antiferromagnetic and ferromagnetic is thus clearly established.

To obtain numerical values for the coupling strength, the data in Fig. 6(a) have been fitted to a theoretical model.[17,18] The calculated dependence of the resonance fields on the interlayer coupling is displayed in Fig. 6(b) by the solid lines. Input parameters are the bulk gyromagnetic ratio of Co (2.18), the microwave frequency at which the experiments have been performed (34.3 GHz), the orientation of the applied field (along the film plane), the products $t_1 M_{s1}$ and $t_2 M_{s2}$ (determined from VSM experiments), and the anisotropy fields of both layers (8.7 and 12.5 kOe) which fix the crossing points with the $J = 0$ axis. These anisotropy fields have been chosen to agree with the experimentally observed resonance fields at large Ru thicknesses. The data of Fig. 6(a) are now inserted in Fig. 6(b) at a vertical position which is determined by the measured resonance field, and at a horizontal position corresponding to optimal match of the lowest resonance field with the lowest H versus J branch. For clarity, the data points (solid circles) are labeled with the corresponding Ru thicknesses. The coupling strength belonging to a certain Ru thickness can now be read off immediately. Before discussing the results, a few comments are in order.

Due to the absence of the optical mode in the AF region and the insensitivity of the acoustic mode to J, the determination of the strength of the AF coupling, in the present case, is extremely sensitive to changes in the 70 Å Co layer's anisotropy field. For example, a minute increase causes a slight shift of the lower curve to smaller resonance fields, but adjustments of the horizontal positions of the data needed to restore optimal match with the changed calculation are considerable. A 4% change in the anisotropy field, for example, results in a change with a factor of 4 of the AF coupling strength at 4 Å Ru. Such small changes in the magnetic anisotropy are likely to occur. For instance, a possible dependence of the roughness of the top Co/Ru interface on the Ru thickness with its accompanying change of the interface contribution to the magnetic anisotropy could account for such deviations.[19] The AF coupling values obtained from these FMR data are therefore unreliable (and will not be used). The ferromagnetic coupling values, however, are fairly insensitive to errors in the anisotropy fields (the error bars resulting from a 4% change in the anisotropy fields are smaller than the plot symbols).

For the sake of completeness and to check the compatibility with series A and B, the AF coupling strengths of the present samples have been determined from VSM measurements. These were performed again with in-plane applied fields. Typical hysteresis loops are displayed in Fig. 7. No magnetic anisotropy is involved here since the layers are grown in the (111) orientation and rotations are confined to the layer planes. This situation is comparable to the $K = 0$ case, to which Fig. 2(c) and Eq. (5) pertain. However, as a result of the unequal layer thicknesses in the present case, the symmetry is broken and the magnetization curve of Fig. 2(c) is modified. Absolute minimum energy calculations yield the magnetization curve shown in Fig. 8(a). The loop is characterized by a plateau at low fields corresponding to an antiparallel alignment of the Co magnetizations, with the one pertaining to the thickest layer parallel to the field, followed by a gradual increase of the magnetization corresponding to a rotation process in which both Co layers participate. From Fig. 8(b), showing the angles of both magnetizations with respect to the field, the detailed behavior during the course of the transition can be seen. The fields H_1 and H_2 at which the rotation of the layer magnetizations commences and ends, respectively, are derived easily from the stability condition and are given by

$$\mu_0 H_1 = -2J\left(\frac{1}{t_1 M_{s1}} - \frac{1}{t_2 M_{s2}}\right), \quad (7)$$

$$\mu_0 H_2 = -2J\left(\frac{1}{t_1 M_{s1}} + \frac{1}{t_2 M_{s2}}\right). \quad (8)$$

Note that for equal magnetic layers H_1 reduces to zero and H_2 reduces to Eq. (5), as expected. We remark here that Eq. (8) was derived also by Heinrich *et al.*[20]

The shape of the experimental hysteresis loops of Fig. 7 is in good agreement with this minimum energy model. For a Ru thickness of 10 Å a ferromagnetic coupling is observed, while the loops obtained for Ru thicknesses of 4, 6, and 16 Å Ru display the typical behavior expected for AF coupling. The AF coupling strengths are deduced using expressions (7) and (8). In Fig. 9 the resulting values are collected together with the ferromagnetic data

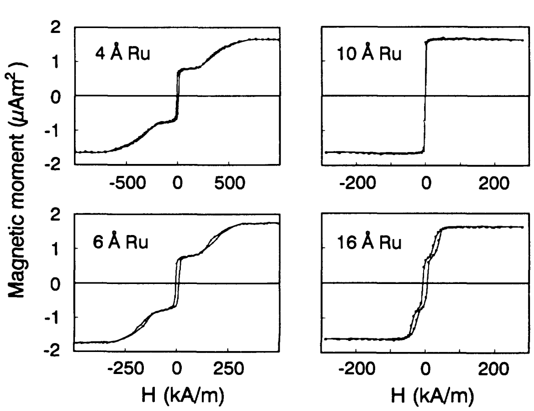

FIG. 7. Hysteresis loops measured by VSM of 200 Å Pd + 30 Å Co/x Å Ru/70 Å Co + 30 Å Pd samples with Ru thicknesses x as indicated. The field is applied along the film plane.

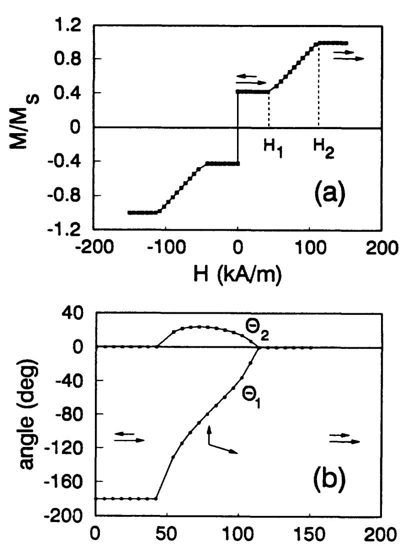

FIG. 8. (a) Theoretical magnetization curve and (b) the corresponding theoretical behavior of the magnetization directions of two AF-coupled ferromagnetic layers having different layer thicknesses and an in-plane preferential orientation. The in-plane angles θ_1 and θ_2 refer to the orientation of the magnetizations relative to the direction of the (in-plane) applied field. These directions are also schematically indicated by the arrows.

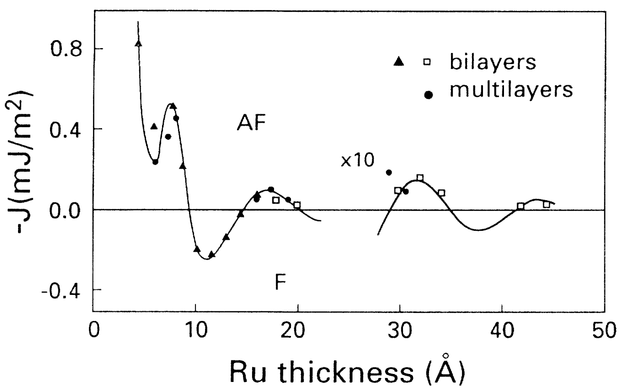

FIG. 9. Interlayer exchange coupling values versus the Ru thickness obtained for various series of samples. The solid lines are guides to the eye. The values for Ru thicknesses larger than 25 Å have been multiplied by 10.

obtained from the FMR experiments and the AF data obtained for series A and B.

C. Discussion

The results collected in Fig. 9 clearly yield a magnetic coupling that oscillates between ferromagnetic and antiferromagnetic. With respect to earlier studies[4,21,22] we note that we were able to reveal an additional maximum in the AF coupling at 44 Å Ru having a very small coupling strength (-0.0035 mJ/m^2). Detection of this peak has been enabled by the significantly smaller Co layer thicknesses which have been used in the present study.

However, the coupling behavior appears to be somewhat irregular; i.e., the peak-to-peak distance is smaller at smaller Ru thicknesses. In particular the peak at 8 Å Ru contributes to this behavior (this peak has also been reproduced by the authors in a different M/Ru/M system where the magnetic layers M consist of Co/Pd multilayers[23]). We remark that this peak has not been observed in previous studies,[21,22] perhaps due to different sample morphologies in those studies through which the peak is washed out due to an averaging with the very large coupling strengths at the more thinner Ru layers. The irregularity in the peak-to-peak distances might be a manifestation of preasymptotic behavior or is possibly a result of additional oscillatory components in the coupling (having different periods, phases, and falloff rates). Recently it has been shown by Stiles that the occurrence of such a multiperiodic coupling for the Co/Ru system is feasible.[24] The average oscillation period that has been deduced from the present experiment is approximately 12 Å. This is in good agreement with two of the periods (11.2 and 11.6 Å) that have been derived by Stiles from the extremal Fermi surface spanning vectors of Ru(0001) (Ref. 24) and is also consistent with the earlier observations that were limited to AF maxima between 10 and 40 Å Ru.[4,21,22]

The present range in coupling strengths which are obtained for the oscillation maxima and which span two orders of magnitude offers the opportunity to make a detailed comparison with the Ruderman-Kittel-Kasuya-Yosida (RKKY) prediction for the envelope of the range function in the asymptotic regime. The latter predicts that, at $T = 0$ K, the amplitude $J_{\max}$ of the oscillatory coupling should decrease quadratically with increasing spacer layer thickness:[25]

$$J_{\max} = J_0/t^2. \tag{9}$$

To compare this behavior with the present data (four AF maxima and one ferromagnetic maximum), Fig. 10 presents a log-log plot of the moduli of the coupling strengths at the maxima as a function of the Ru thickness. The straight line that has been included has a slope corresponding to the quadratic decrease and an offset which had been adjusted via J_0. It is seen that the first three maxima obey the quadratic behavior, whereas the coupling at larger Ru thicknesses seems to fall off faster. The origin of the latter behavior might be a result of the fact that the experiments were performed at $T = 300$ K rather than at 0 K. Recent theories that take into account the "smearing of the Fermi surface" associated with the more gradual variation of the Fermi-Dirac distribution function at finite temperatures have predicted that the temperature dependence of the exchange coupling is stronger for larger interlayer thicknesses.[26,27] Alternatively, at finite temperatures the deviations from the $1/t^2$ law will be larger for the thicker interlayers. In order to infer if the *magnitude* of the present deviations is physically realistic within the above-cited theories, the following expression has been fitted to the data:[26,27]

$$J_{\max} = (J_0/t^2)\frac{Tt/\alpha}{\sinh(Tt/\alpha)}. \tag{10}$$

Here, α is a parameter depending on the Fermi velocity at the Fermi surface extremum which corresponds to the oscillation period.[26,27] The resulting fit, which was obtained by variation of J_0 and α and which is shown as a

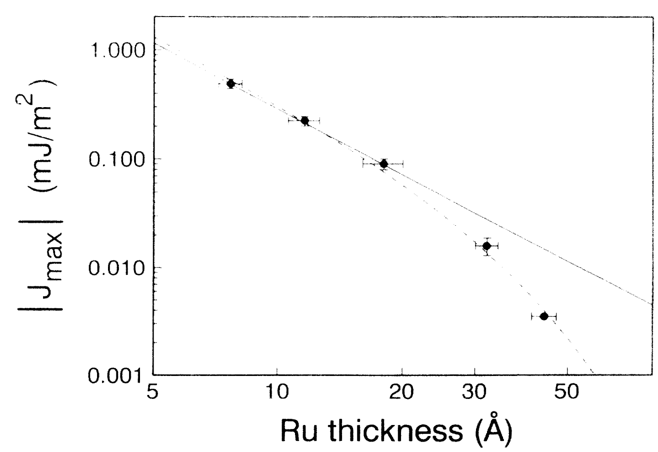

FIG. 10. Log-log plot of the moduli of the coupling strengths as a function of the Ru thickness. The solid line represents a quadratic decrease, whereas the curved line represents a fit including a parameter which accounts for the temperature dependence of the coupling.

curved line in Fig. 10, yields $\alpha = (3.9 \pm 0.3) \times 10^3$ Å K. Unfortunately, no detailed predictions are yet available for this parameter for Ru(00.1) spacers. However, the above value is comparable in magnitude to the RKKY prediction for Cu(111) which yielded $\alpha = 4.8 \times 10^3$ Å K.[26] The value is also comparable to an experimental result obtained for Ru: $\alpha = (1.92 \pm 0.06) \times 10^3$ Å K.[28]

Although the Fermi surface smearing at room temperature as discussed above could account for the experimental observations, effects due to reductions in the electron mean free path resulting from defects in the spacer layer might also be the cause for the observed behavior. As discussed by de Gennes,[29] such a decrease in mean free path decreases the amplitude of the oscillations. However, a fit of $t_{\mathrm{Ru}}^{-n} \exp(-t_{\mathrm{Ru}}/l)$ as suggested by de Gennes seems to give unrealistic parameters. A mean free path l of 0.9 Å only and $n = 1$ have been obtained. A discussion, in which the layered geometry of the problem has been included, has been given by Bruno.[26] Here, a certain cutoff thickness above which the coupling should decrease fast is defined. Since it is determined by the unknown average distance between the lateral imperfections (dislocations, grain boundaries, and thickness fluctuations) and the presently unknown angle subtended by the Fermi velocity of the electrons "which carry the coupling" and the film plane, a discussion within this model is precluded.

To conclude this section, a comparison is made between the magnitude of the coupling strengths of the present work and the literature. Large discrepancies are concluded. For the second AF peak, which is the first maximum that allows comparison, coupling strengths differ by a factor 1.5–5 (Refs. 4, 21, and 22 and the present work). This indicates that the preparation method is very important. The different preparation conditions yield possibly a different interface roughness and microstructure, affecting the various magnetic properties and the resulting magnetic behavior. The latter can be demonstrated by a comparison of the present hysteresis loops and loops obtained for evaporated samples.

Figure 11 shows the hysteresis loops of a 10×(11 Å Co/8 Å Ru) multilayer grown by *evaporation*[15] and by *sputtering* (present work). Clearly both loops exhibit AF coupling; however, the zero susceptibility at low fields present for the sputtered sample is certainly not observed for the evaporated sample. This absence of zero susceptibility was also observed by Ounadjela *et al.*[21] for MBE-grown Co/Ru samples and was proposed to be due to an inhomogeneous rotation mechanism for spins *within* the Co layers as a result of competition between the supposedly strongly reduced bulk Co *intralayer* exchange and the Ru interlayer exchange. However, as stated already, the nonzero susceptibility might also have a microstructural origin. Transmission electron micrograph (TEM) experiments performed on both samples of Fig. 11 revealed a significantly different microstructure for the evaporated sample compared to the sputtered sample. For the sputtered sample the layers appeared flat and continuous across the grain boundaries which were at least 500 Å apart. For the evaporated sample on the contrary, the crystallite size was considerably smaller (≈ 200 Å), a significant bending of the layers occurred, and continuity of the layers across the grain boundaries could not be concluded. A strong reduction of the perpendicular magnetic anisotropy localized near the grain boundary is possible and could account for the nonzero susceptibility in our case.

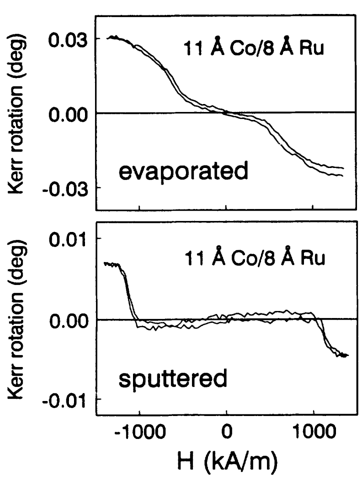

FIG. 11. Hysteresis loops of a 10×(11 Å Co/8 Å Ru) multilayer grown by evaporation (Ref. 15) and by sputtering.

VI. CONCLUSIONS

Results on the magnetic interlayer coupling in sputtered Co/Ru bilayers and multilayers have been presented. These have been derived from a combined MOKE, VSM, and FMR study.

The interlayer exchange coupling has been found to oscillate as a function of the Ru thickness with a period of approximately 12 Å. This is in agreement with a recently predicted set of possible oscillation periods, including 11.2 and 11.6 Å,[24] and also agrees with previous experiments.[4,21,22] The observed coupling behavior was somewhat irregular; i.e., the peak-to-peak distance at small Ru thicknesses was considerably smaller than for larger spacer layer thicknesses. This might be a signature of preasymptotic behavior possibly combined with the presence of additional oscillatory components in the coupling. The strength of the coupling was found to decrease as t_{Ru}^{-2} up to a Ru thickness of about 20 Å above which deviations from this quadratic behavior occurred. These might be explained by the fact that a stronger temperature dependence of the coupling is expected with increasing interlayer thickness.

The hysteresis loop experiments in themselves yield the following conclusions: The hysteresis loops of the AF-coupled bilayers with a perpendicular preferential orientation displayed very clearly the expected spin-flip transitions — this in contrast to earlier observations on MBE-grown samples[21] and high vacuum evaporated samples,[15] which exhibited less well-defined spin-flip fields. The magnetization behavior of the present Co/Ru *multilayers* displayed *two* transitions which were *independent* of the number of magnetic layers and are likely related to the fact that layers at the boundary of the multilayer stack are coupled only once and therefore reverse their direction at an other (lower) applied magnetic field than the inner layers which are coupled twice.

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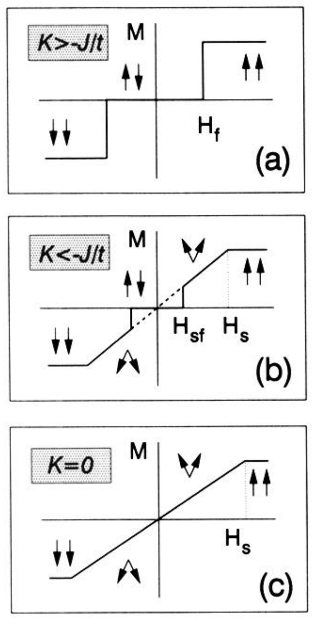

FIG. 2. Theoretical magnetization curves for two identical AF-coupled ferromagnetic layers having a uniaxial magnetic anisotropy which is (a) larger than the coupling strength and (b) smaller than coupling strength. The arrows schematically indicate the orientations of the magnetizations relative to the vertical easy axis, along which the field is applied. The situation of zero anisotropy (c) is a special case of (b).