Antiferromagnetic coupling in (111)-oriented Co/Pt superlattices

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(Received 25 January 1994)

Nuclear magnetic resonance (NMR) and low-temperature magnetization measurements are used to demonstrate antiferromagnetic (AF) coupling in molecular beam epitaxy grown (111)-oriented Co/Pt superlattices. Drastic changes in the NMR spin-echo spectra as a function of the in-plane dc magnetic-field strength provide conclusive evidence of AF interlayer coupling in these structures, consisting of 35- and 52-Å Co and 18-Å Pt bilayers. The structure of the Co films is mainly hcp (70%) and the coupling energy per unit area, as determined from in-plane magnetization loops is found to range up to 1 erg/cm^2.

I. INTRODUCTION

Since the discovery of the oscillatory exchange coupling[1] between magnetic layers via a nonmagnetic spacer layer, a great number of transition-metal (TM) spacers have been explored.[2] The periods of exchange oscillations are determined by the geometry of the Fermi surface of the TM spacer.[3] The interest of magnetic films exhibiting interlayer antiferromagnetic (AF) coupling resides in their eventual giant magnetoresistance (GMR).[4] GMR occurs because of the drastic difference in the resistance of the film between the antiparallel and parallel (in a saturating field) alignments of the magnetic layers. This reflects the fact that the electron mean free path in a ferromagnetic transition metal is spin dependent. The strength of the AF coupling is found to increase exponentially with increasing number of spacer d electrons. However, surprisingly enough, Co/Pt multilayers (Pt has $5d^9$ configuration) were reported to exhibit no AF coupling.[2] The reason may be that AF coupling is obscured by magnetic anisotropy or interface roughness.[2] Very recently the periods of exchange oscillations for (111)-oriented Pt spacer layers were calculated by Stiles[5] to be either short ($\sim$5 Å) or long ($\sim$25 Å), as compared to usual values in the range 9–12 Å. In this paper we report spin-echo nuclear magnetic resonance (NMR) and magnetization studies in (111)-oriented Co/Pt multilayers containing thick Co layers, prepared by molecular-beam epitaxy with Pt layer thickness around 18 Å. Conclusive evidence for AF coupling in these samples is found both from the field dependence of NMR spectra and magnetization experiments.

II. SAMPLE PREPARATION AND CHARACTERIZATION

The samples with Co layer thicknesses of 35 and 52 Å were grown on (0001) sapphire and (111) GaAs substrates, respectively. The Pt and Co layers are parallel to the (111) plane. Both samples contained 15 Co/Pt bilayers with Pt layer thickness of about 18 Å. The multilayers were grown using different seed films. A 200-Å-thick Ag(111) film was used for growth[6] on GaAs(111) and a 30-Å-thick Pt(111) film for growth[7] on sapphire (0001).

High-angle x-ray specular scans for both samples are shown in Fig. 1. For the sample grown on sapphire (sample 1) a set of six superlattice peaks is observed, overlapping a broad peak due to the Pt seed layer. The Al_2O_3(0006) peak is extremely sharp and was not recorded in this case. In the scan for the sample grown on GaAs(111) (sample 2), a big Ag(111) peak showing total thickness fringes can be observed. This peak extends through the superlattice diffraction region, some of which satellites appear strongly distorted.

The experimental data for sample 1 were fitted using a parameter refinement procedure.[8–10] Bulk in-plane and out-of-plane spacings were assumed for the Pt film. NMR measurements indicate that about 30% of the Co films is fcc in both superlattices, and a weighted average of hcp and fcc Co interplanar spacings was therefore assumed, taking the hcp in-plane atomic density (equivalent to the fcc one within 0.1%). Bulk Pt and hcp Co Debye-Waller and scattering factors were assumed. Layer thickness fluctuations are taken into account by assuming a discrete Gaussian variation of the number of atomic planes in each layer about an integer average value N. The width of the Gaussian distribution is denoted by σ_N. Chemical interdiffusion can be added to the calculation by substituting the scattering factors (fCo and fPt) of both materials in the layers at the interface by a weighted average of both scattering factors with an interdiffusion parameter $v(1)$. In this way, the scattering factor of the first Co layer is given by $[1-v(1)]f\mathrm{Co}+v(1)f\mathrm{Pt}$. In the second and third monolayers the interdiffusion parameter is given by $v(1)\exp(-v(2))$ and $v(1)\exp(-2v(1))$, respectively. Finally, the distance between the last layer of a material and the first of the other one can be varied using a Gaussian distribution of width $v(3)$ about the average value $[(d\mathrm{Co}+d\mathrm{Pt})/2]$. The contribution of the Pt(111) seed layer was included using a Lorentzian peak. Figure 1(a) shows experimental data and the best fit ob-

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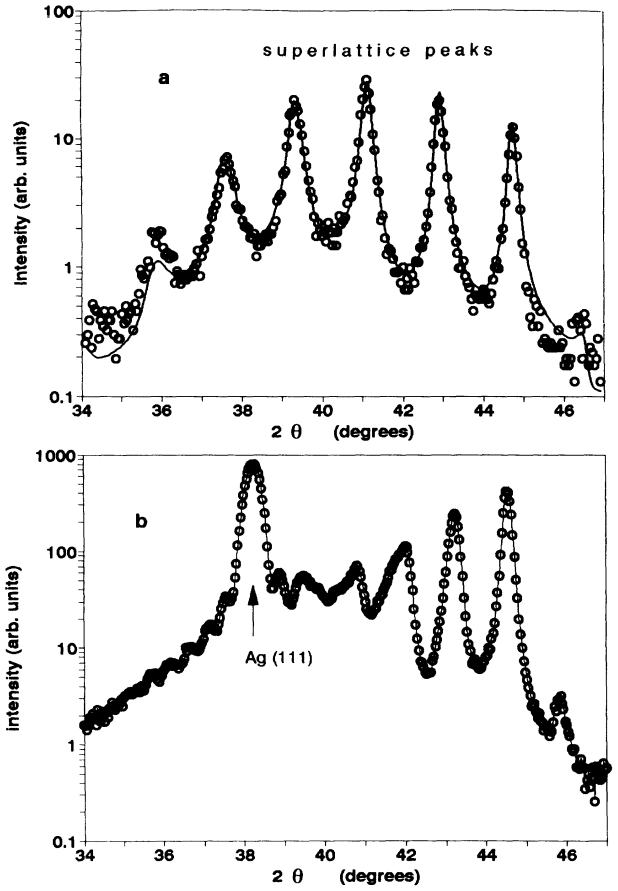

FIG. 1. High-angle x-ray-diffraction results. (a) $Al_2O_3(0001)/30$ Å Pt/[35 Å Co/18 Å Pt]×15. The solid line is a fit to the experimental data as explained in the text. (b) GaAs(111)/200 Å Ag/[19 Å Pt/52 Å Co]×15/19 Å Pt. The Ag(111) contribution with total thickness fringes can be observed in the left side of the scan.

tained. From the fit the Co and Pt thicknesses are found to be 34 and 19 Å, respectively. The thickness distribution within each layer is summarized in Table I.

The strong contribution of the Ag(111) peak in sample 2 makes a similar analysis difficult to realize. Nevertheless, a study restricted to the peaks far from the Ag peak allows us to estimate the Co and Pt layer thicknesses to be 52 and 19 Å, respectively. No direct information concerning interfacial roughness can be extracted. Since both samples were grown under the same conditions it is possible, in principle, to extend some of the results obtained for sample 1 to sample 2. Fitting the experimental data of this sample using as input parameters the numbers obtained for sample 1, a broader thickness distribution can be concluded, extending up to 9 and 4 monolayers (ML) wide in the Co and Pt layers, respectively. One should be, nevertheless, very conservative about these numbers. Previous high-resolution transmission-electron-microscopy (HRTEM) studies on Co/Pt superlattices grown under the same conditions[7] showed an extensive growth disorder, specially twins and stacking faults, that could explain this thickness dispersion. The distortion of several superlattice peaks near the Ag(111) is noticeable in sample 2. One could interpret such a distortion as an overlapping of the superlattice peaks with the total thickness fringes coming from the Ag, but neither their intensities nor their positions are in agreement with such an assumption. An effect due to the coexistence of fcc and hcp Co can be a tentative explanation: if the grain size for fcc and hcp Co in sample 2 is big enough, coherent diffraction from both phases can happen and the results may be interpreted as the contribution from two different superlattices, one with Co fcc and the other with Co hcp. The overlap of both contributions would give rise to the observed distortion in the peaks. Since the Co layers in sample 1 are thinner, the smaller fcc and hcp Co grains would give rise to an average Co lattice constant instead, with a single contribution to the diffraction process.

TABLE I. Thickness distribution within the Co and Pt layers.

	Thickness (Å)	N (ML)	σ_N	$v(1)$	$v(2)$	$v(3)$
Co	35±2	17	0.99	0.02	7.9	0.16
Pt	18±0.7	8	0.26			

III. EXPERIMENTAL RESULTS AND INTERPRETATION

The Co NMR measurements were carried out at 2 K using a variable-frequency spin-echo apparatus. The sample was mounted inside a Dewar tail around which was fitted an exciting coil, such that an rf field, of up to about 12 Oe, was parallel to the film plane. A dc magnetic field of up to 12 kOe can be applied perpendicular to the coil axis, either in plane or perpendicular to the sample surface. For ^{59}Co nuclei, a shift ΔH (kOe) in the resonance field is related to a shift ΔF (MHz) in the resonance frequency by the ratio $\Delta F/\Delta H = 1.0054$ MHz/kOe.

A broad Co spin-echo spectrum in zero external field as viewed in the top of Fig. 2 is characteristic of hcp Co.[11] The most important information, however, is contained in the continuous change of the line shapes in the presence of an increasing dc magnetic field parallel to the film plane. The magnetization is lying in the film plane for the present samples with Co layer thicknesses equal or greater than 30 Å. When the dc field is increased to 4 kOe a new sharp line appears in the low-frequency wing. This line is not related to an in-plane anisotropy as might be expected since the NMR measurements were unchanged after rotating the films 90° about the normal direction. Similar results are obtained with the sample with Co layer thickness of 35 Å. To understand the presently observed shape transformation better, we first compare the present field-induced spectra of Co/Pt multilayers with spectra observed in zero external field, in a (111)-oriented Au/70 Å Co/Au sandwich[11] (dashed line in top panel). The sharp and broad lines were shown to arise from the fcc and hcp phases, respectively. In a dc magnetic field parallel to the film plane these lines are simply shifted, without any deformation to lower frequency as the hyperfine field H_n is negative. The field

dependence of the resonance spectra in the present Co/Pt samples is interpreted as evidence for AF coupling in these samples and is discussed as follows.

First it is recalled that the spin-echo signal in magnetic materials is proportional to the enhancement factor η of the rf field. In ferromagnetic films the factor η, which results from domain rotation, is equal to the ratio H_n/H_A (Ref. 12) where H_A is the in-plane anisotropy field. For (111) texture the H_A value is checked to be small as evidenced from torque measurements. In antiferromagnetic films with negligible in-plane anisotropy, η is determined by the AF coupling strength. To show this we assume that a field H is applied, in the plane, perpendicular to the antiferromagnetic direction. The magnetization M_s will deviate by an angle θ from this direction. The energy density function, neglecting the magnetic anisotropy, can be written as

$$E=-HM_s\sin\theta-(J_{AF}/t)\cos2\theta\ , \tag{1}$$

where J_{AF} is the interlayer exchange parameter per surface unit and t the magnetic layer thickness.

The equilibrium angle θ is given by the relation

$$\frac{\partial E}{\partial\theta}=-HM_s\cos\theta+(4J_{AF}/t)\sin\theta\cos\theta=0$$

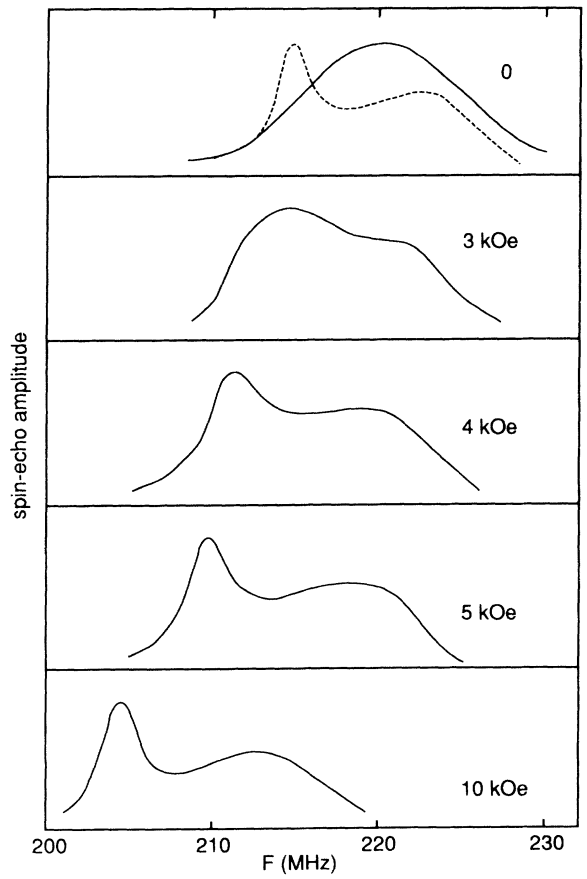

FIG. 2. Co spin-echo spectra at 2 K in Co/Pt superlattice with Co layer thickness of 52 Å, for different values of magnetic field parallel to the film plane. The dashed line represents the spectrum in a (111)-oriented Au/70 Å Co/Au sandwich exhibiting a sharp fcc and a broad hcp line.

or

$$\sin\theta=\frac{HM_st}{4J_{AF}}\ . \tag{2}$$

If H is a rf field the nuclei will see an effective transverse rf field $H_n\sin\theta$. The enhancement factor is defined as

$$\eta=\frac{H_n\sin\theta}{H}=\frac{H_n}{H_{AF}} \tag{3}$$

with

$$H_{AF}=\frac{4J_{AF}}{M_st}\ .$$

From relation (2), H_{AF} is identical to the saturation field. If the multilayer contains regions which experience strong AF coupling but are only loosely ferromagnetically coupled with other regions, η will vary locally within the sample. As a consequence only the signal with high η values can be observed in zero external field. This signal is strongly reduced in a dc magnetic field greater than H_A. Consider now a dc field H applied in the plane in any direction, stronger than the flip field to make all the antiferromagnetic directions perpendicular to H. Applying a *small* field h perpendicular to H is equivalent, in first order, to simply rotate H by an angle α given by $\tan\alpha=h/H$. Consequently, the magnetization and the hyperfine field are also rotated by α. The rf field is several orders of magnitude smaller than the dc field so that $\tan\alpha\sim\sin\alpha\sim\alpha$. In this case the enhancement factor is given by

$$\eta=\frac{H_n}{H}\ , \tag{4}$$

which is the same as for a ferromagnetic film, with $H\gg H_A$. Thus in a saturating field the resonance spectra reflect the actual distribution of the hyperfine field. Once the saturation is reached there is no further deformation of the spectra as for a simple ferromagnetic film. The fraction of the fcc phase, represented by the sharp line observed in a saturating field, is estimated to be 30%.

The present NMR findings are corroborated by magnetization measurements. The in-plane magnetization curve shown in Fig. 3 is typical of multilayers with AF coupling or of ferromagnetic films having an in-plane uniaxial anisotropy with the c axis perpendicular to the field. This curve is independent of the orientation of the field within the plane, supporting the AF behavior by ruling out an anisotropy effect. The remanent magnetization of about 15% represents the fraction of the sample which experiences no AF coupling. The convex shape of the magnetization curve reveals a distribution in AF coupling strength. Within the approximation of negligible in-plane anisotropy, the saturation field H_s is identified as the maximum H_{AF} value. The in-plane magnetization curve can be fitted by assuming a distribution of H_s values ranging from zero (ferromagnetic fraction) to a maximum value. Good agreement is obtained by assuming, for the AF regions, equal contributions with

H_s=2,3,4,5,6 kOe for the sample with 52-Å Co layer thickness. There is a close correspondence between the NMR and magnetization results. The NMR signal in zero external field arises mainly from regions showing remanent magnetization. In a dc field of a few kOe the spread in the enhancement factor across the sample is reduced such that the signal due to AF regions can be observed. The resolution of the fcc lines is achieved at the saturation field, where all the fcc hyperfine fields are antiparallel to H. Similar behaviors were observed for the sample with 35-Å Co layer thickness with a maximum H_s value of 8 kOe.

The maximum J_{AF} value is about 1 erg/cm^2 for both samples. The mechanism of AF coupling may be the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction at large distance which has been shown to overcome the d-d interaction between the central Pd layer and the Co layers in Co/Pd multilayers.[13] The respective d-d interaction which leads to the well known ferromagnetic polarization of Pt is smaller for Pt atoms so that the RKKY interaction is expected to be more apparent in Co/Pt multilayers.

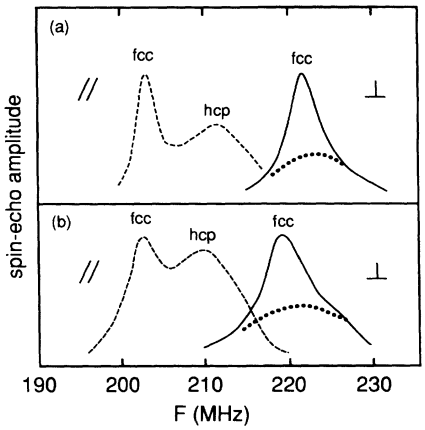

FIG. 4. Co spin-echo spectra at 2 K in a field of 11 kOe perpendicular (solid lines) and parallel (dashed lines) to the film plane for the samples with Co layer thicknesses of (a) 52 Å and (b) 35 Å. The dotted lines represent the expected contributions of hcp Co.

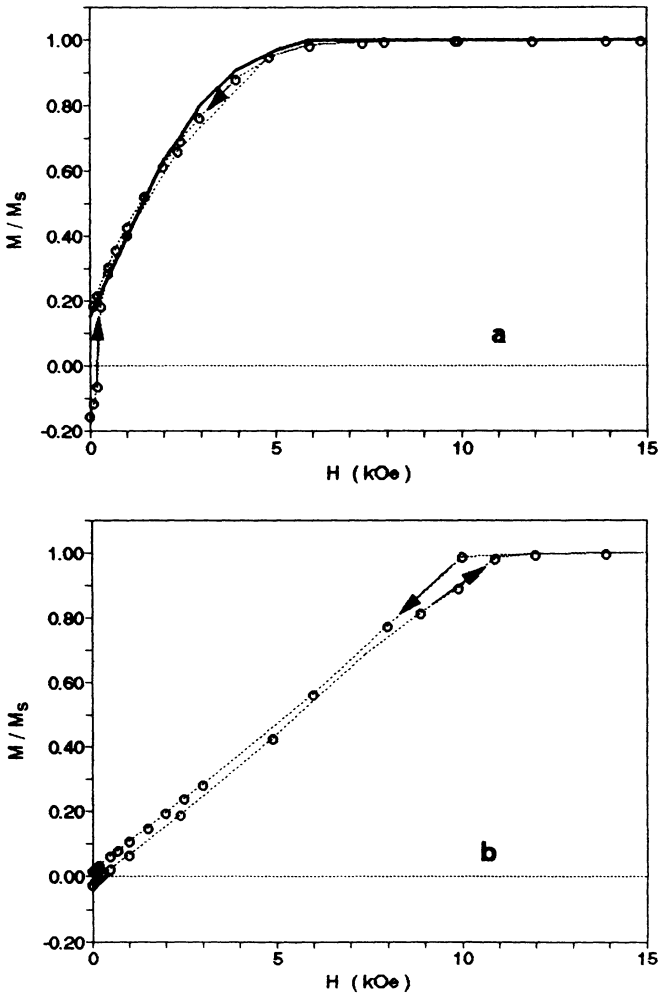

FIG. 3. Reduced magnetization at 10 K as a function of dc magnetic field parallel (a) and perpendicular (b) to the film plane for the sample with Co layer thickness of 52 Å. The solid line represents the calculated curve assuming a distribution of AF coupling strength with H_s values equal to 2, 3, 4, 5, and 6 kOe, respectively.

The hyperfine field is expected to be quasi-isotropic for the fcc phase and anisotropic for the hcp one.[14] With respect to the parallel polarization the resonance frequency of the fcc line in perpendicular orientation should increase by about 18 MHz, due to the demagnetizing field $4\pi M \sim 18$ kOe of Co. For bulk hcp Co, the hyperfine field with magnetic moment along the c axis has been reported to be 8 kOe smaller than that associated with the moment lying in the c plane.[15] Thus for (0001) Co layers the resonance frequency in the perpendicular orientation is expected to increase by 10 MHz. This is born out by our results as shown in Fig. 4. We measure fcc shifts of 16.5 and 18.5 MHz for the samples with Co layer thicknesses of 35 and 52 Å, respectively. Note that a tensile strain exerted from the Pt lattice onto the Co layers will slightly distort the fcc Co lattice resulting in a reduced shift. This strain is expected to decrease as the Co thickness increases, showing better agreement for the 52-Å sample. For the hcp spectra the shifts are estimated to be 11±1 MHz, showing that the anisotropic part of the hyperfine field is of the order of the bulk value.

IV. CONCLUSION

In conclusion, AF coupling in (111)-oriented Co/Pt superlattices with the Pt layer thickness of 18 Å has been observed by the simultaneous use of the spin-echo method and magnetization results. The coupling energy per unit area is found to range up to 1 erg/cm^2. The structure of the Co films is mainly hcp (70%), as deduced from NMR spectra. This proportion is coherent with the high-angle x-ray studies which also imply that fcc and hcp Co phases arise from different crystallites. These results confirm the model, used in this paper, of separate regions exhibiting local interlayer AF coupling of different strengths which appear through the analyses of the NMR and magnetization data.

ACKNOWLEDGMENTS

We thank M. Toney for help with x-ray characterization and Professor I. K. Schuller for providing the x-ray refinement program which was developed with funds provided by the U.S. Department of Energy and the Belgian Interuniversity Attraction Pole Program. The Institut d'Electronique Fondamentale is Unité de Recherche Associée au Centre National de la Recherche Scientifique No. 22.

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