PHYSICAL REVIEW B VOLUME 45, NUMBER 21 1 JUNE 1992-I

Structural study of cobalt-copper multilayers by NMR

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(Received 9 December 1991)

With this NMR study, we show that the quality of the growth of Co/Cu multilayers depends significantly on the Co- and Cu-layer thicknesses: The initial stacking of Co layers is fcc but the hcp stacking tends to develop with increasing Co-layer thickness. On the other hand, the thicker the Cu layers are, the better the fcc growth is. By a comparison of the NMR spectra with those expected from model interface structures, we show that the interfaces between copper and thin Co layers can be described as follows. If the presence of defects in the bulk of the Co layers is excluded, the NMR spectra observed in samples with thin Co layers must be explained by an intermixing of Co and Cu over three monolayers. However, such a model does not hold true for samples with thick Co layers where the existence of bulk defects is strongly suggested. In such a case, the spectra for all the samples studied can be explained by sharp Co/Cu interfaces, as in UHV-grown multilayers, and about 0.7% impurities in the bulk of the layers or possibly columnar grains with an in-plane diameter of about 90 Å. Although model-agreement factors tend to favor the second model (sharp interfaces and bulk defects) rather than the first one (diffused interfaces), additional information on the nature of the bulk defects is needed.

I. INTRODUCTION

Currently multilayers of ferromagnetic slabs separated by nonmagnetic metals are very widely studied as they present peculiar magnetic properties including, for various magnetic-nonmagnetic couples, the existence of giant magnetoresistive effects.[1-6] From this point of view, cobalt-copper multilayers are particularly interesting as they exhibit an oscillating antiferromagnetic-ferromagnetic exchange coupling, which oscillates as a function of Cu thickness, and very large saturation magnetoresistances as high as 65% at 300 K.[3-6] Such magnetoresistive effects as well as the oscillating magnetic coupling between ferromagnetic layers are found to depend on the fabrication process of the multilayers. To get further understanding of the magnetic and magnetotransport properties, structural studies of the multilayers and particularly of the interface nanostructure are needed to establish possible correlations between the structure of the multilayers and their properties. Previous NMR studies[7] have demonstrated the ability of the technique to shed some light on the structure and composition of the interfaces at the nanoscopic scale. It is the aim of this paper to report on such an NMR study of the structure of sputtered cobalt-copper multilayers, which have been shown to present strong magnetoresistive effects.

After a presentation of the experimental method which we use, including the measurement of bulk Co-Cu alloys for reference (Sec. II), the paper proceeds to a coarse analysis of the spectra in terms of proportion of cobalt atoms in fcc and hcp environments or in intermixed regions (Co/Cu interfaces) (Sec. III). The last section of the paper is devoted to a more quantitative comparison of the experimental spectra with simulated spectra according to various models of the interface structure.

II. EXPERIMENTAL

^{59}Co NMR has been performed on Co/Cu multilayers produced by sputtering onto chemically etched Si ⟨100⟩ wafers kept at 0 °C during deposition. For each sample, after deposition of a first Cu or Fe buffer layer, 30 Co/Cu bilayers were grown with the same sputtering conditions as those used for the data of Ref. 6. The respective thicknesses of Co and Cu layers are given in Table I.

The multilayers were characterized by x-ray diffraction.[8] The periods determined from the distance between the high-angle satellite peaks of the x-ray data are always in good agreement with the nominal thicknesses ($\pm 2\%$). Those measurements also show a ⟨111⟩ texture of the multilayers with a rocking curve about 10° wide. For 15-Å Co thickness series the coherence length along the growth direction is about 400 Å, which corresponds to one-third of the total thickness of the multilayers.

A spin-echo NMR experiment in zero field was performed using an automated frequency scanning broadband spectrometer with phase coherent detection. For maximum sensitivity, the NMR observations were performed at 1.5 K. Each spectrum presented in this paper is the result of a set of 5–7 experimental spectra (each one comprising 251 frequency spots) obtained for different strengths of the radiofrequency field H_1. Since, in ferromagnetic materials, H_1 and the NMR signal are

TABLE I. Characteristics of the measured multilayers.

Sample	Co thickness (Å)	Cu thickness (Å)	Buffer
(15-Å Co)/(15-Å Cu)	15	15	Fe
(15-Å Co)/(20-Å Cu)	15	20	Fe
(60-Å Co)/(20-Å Cu)	60	20	Cu
(60-Å Co)/(60-Å Cu)	60	60	Cu
(60-Å Co)/(90-Å Cu)	60	90	Cu

mediated (and enhanced) by the rotation of the local electronic moments, it is a general rule for us to perform several spectrum observations for different H_1 strengths ranging over more than one order of magnitude. At each frequency, this provides a measure of the enhancement factor $\eta(\omega)$ resulting from the electronic susceptibility; in turn it allows one to correct the observed NMR intensity for this enhancement factor beside the usual ω^2 dependence of the NMR signal. Causes for a frequency dependent η include the frequency-dependent initial susceptibility of the material. In addition, for inhomogeneous materials (e.g., phase admixture), different regions of the sample giving rise to different NMR lines may have different electronic magnetization stiffness and hence different η's.[9] This is the case, in the present study, between lines arising from Co nuclei sitting in the bulk of the Co layers and those from Co nuclei in the Co/Cu interface area. Such a procedure enables us to get a quite accurate density of nuclei per frequency, which is needed for structural investigations.

The basis for the use of NMR in structural investigations of ferromagnetic material lies in the fact that the hyperfine (hf) field experienced by a nucleus, hence its NMR frequency, is strongly dependent on the nature and number of atoms (or moments) in its neighborhood and possibly on the symmetry of this neighborhood. However, it is quite hard, if not impossible yet, to achieve *ab initio* calculations of hf fields for given atomic configurations of elements in a solid material and particularly in metallic alloys. Therefore, one has to rely on reference measurements on samples of known structure and composition which are hopefully close to the one under investigation. Thus, in order to analyze the spectra in terms of local structure, we have first measured bulk Co-Cu alloys as references. Similar measurements already reported in the literature[10] disagree slightly with each other and this made new measurements necessary. Besides we like to have reference spectra observed within the same experimental conditions, setup, and procedure.

Three alloys with different copper concentrations were prepared (2%, 6%, 10%). They were obtained by quenching from high temperature to stabilize as far as possible an fcc $Co_{1-x}Cu_x$ solid solution despite the low miscibility of the two metals at low temperatures. The NMR spectra of the three alloys are given in Fig. 1 where the development of satellite lines on the low-frequency side of the pure fcc main line can be observed as Cu content increases. From the concentration dependence of the relative intensities of the main line and satellites we can analyze those spectra, in a rather usual way, as follows. The most intense line at high frequency is attributed to cobalt atoms with a local environment of 12 Co nearest neighbors; this line always peaks at about the same frequency as in pure fcc cobalt (217 MHz); its frequency may slightly decrease as the concentration of nonmagnetic element increases, which is understood in terms of the average magnetization of the sample decreasing (long-range effects). The next line, about 16 MHz lower in frequency, is attributed to Co with 11 Co and one Cu nearest neighbors as its relative intensity increases linearly with Cu content. The third line, 16 MHz lower again, the intensity of which increases in a rather parabolic way, is assigned to Co with ten Co and two Cu nearest neighbors. A similar trend is observed in other bulk Co-based alloys (with Ru, Cr, Fe) that we have studied prior to the NMR investigation of the corresponding multilayers (more details will be given in a forthcoming paper).

For the following analysis of the interfaces in Co/Cu multilayers, we shall assume that the neighborhood of cobalt atoms has the same influence on the hyperfine field as in the case of Co-Cu alloys: Each additional Cu atom among the 12 neighbors of a Co nucleus decreases its resonance frequency by an amount close but not necessarily equal to 16 MHz. Indeed, as mentioned above, the symmetry of the neighborhood can also affect the hf field and, for example, three Cu neighbors at random among the 12 neighbors of Co (solid solution case) may not yield exact-

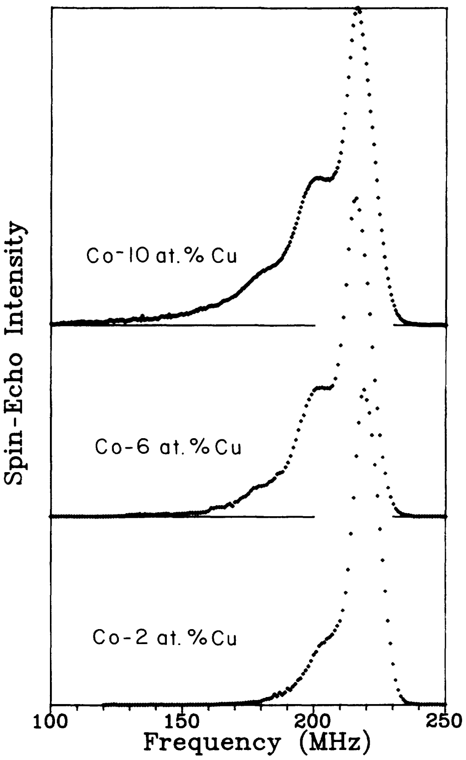

FIG. 1. ^{59}Co NMR spectra of cobalt-copper alloys with 2, 6, and 10 at. % of copper. The main line is attributed to Co with 12 Co nearest neighbors and each successive satellite to the successive substitution of Cu for Co in the vicinity of Co.

ly the same average hf field as three Cu neighbors lying in the same plane of a ⟨111⟩ textured epitaxial superlattice. Our previous study of such an epitaxial sample grown by UHV evaporation[7] has shown that interface satellites are separated by about 19 MHz.

III. EXPERIMENTAL RESULTS ON MULTILAYERS

The Co NMR spectra of the 60-Å Co thickness series are shown in Fig. 2, and those for 15-Å Co thickness in Fig. 3.

As the growth of Co on Cu favors fcc stacking, we expect and observe that the main line of the spectra always peaks around 217 MHz. Yet, in the thick Co layer series, shoulders to this main line can be observed on the high-frequency side, which must be attributed to hcp stacking faults in fcc cobalt[11] and/or hcp Co grains; this is a situation which is commonly encountered even in pure fcc Co because of the martensitic transition between fcc and hcp structures. Actually, in fcc Co, it is possible to identify three different hcp-like lines due to stacking defects (Fig. 4), each one with a slightly different hyperfine field; these three lines collapse into a broad asymmetric line in the same frequency range for pure hcp cobalt. In the multilayers, because of the line broadening, we only manage to distinguish at most two shoulders. The assignment of these shoulders to an enhanced hf field of Co at the interfaces is discarded, as they are not observed in the thin Co layer series for which the interface signal must be relatively stronger; in addition, the study of the bulk alloys show that the vicinity of copper depresses the Co hf field.

On the low-frequency side of the main line, a broad tail is observed in the thick Co samples, which is attributed to regions of the multilayers where Co and Cu intermix and the Co hf field is consequently lowered.

In contrast with the thick Co layer series, spectra observed for the thin Co layer series (Fig. 3) essentially exhibit an fcc main line with no obvious high-frequency shoulder. Correspondingly, the interfacial part of the spectra is rather well structured so that it is possible to distinguish several satellite lines.

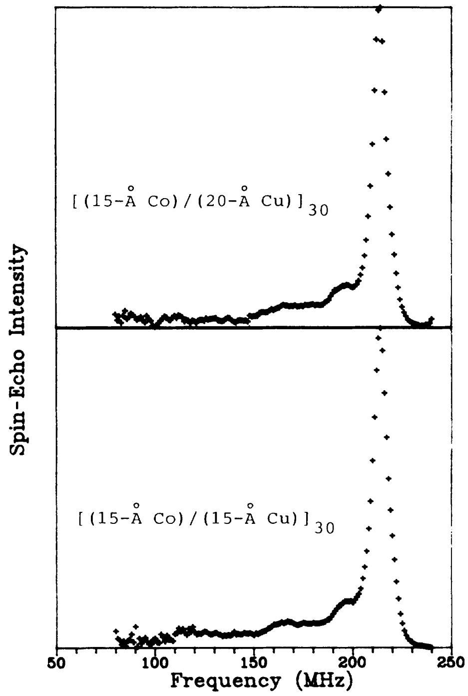

FIG. 3. NMR spectra of the multilayers with 15-Å cobalt thickness. The main line is attributed to fcc Co and the satellites to Co at the interfaces of the multilayers.

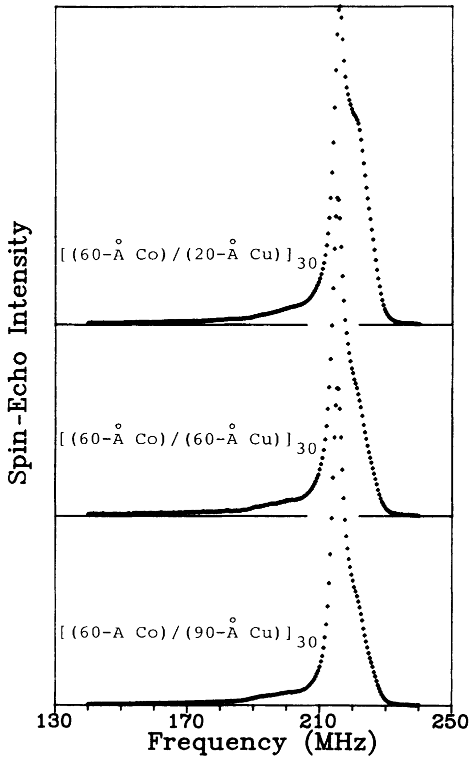

FIG. 2. NMR spectra of the multilayers with 60 Å of cobalt. The main line is attributed to fcc Co, the two upper lines to hcp Co, and the satellites at low frequencies to Co at the interfaces of the multilayers.

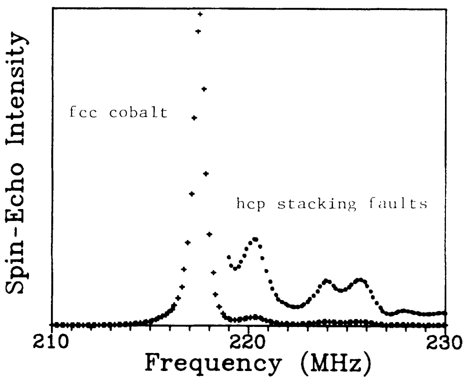

FIG. 4. fcc cobalt NMR spectrum showing hcp stacking faults.

These observations show that any spectrum can be separated into three parts: (1) a set of two uppermost lines at 220 and 225 MHz, which are attributed to Co nuclei in a pure Co neighborhood with hcp-like stacking; (2) then, around 216 MHz, a well-defined single line, which corresponds to pure fcc Cobalt; and (3) the lowest part of the spectra (below 205 MHz), which corresponds to Co nuclei that have less than 12 Co neighbors (and correspondingly a reduced hyperfine field). This is tentatively attributed to Co atoms located in the mixed interfaces between cobalt and copper layers (this attribution will be critically discussed in the next section). Hereafter we use the term interfacial cobalt atom for any Co which belongs to a mixed monolayer or which has at least one Cu nearest neighbor. This definition takes into account all possible cases of interface structure from a perfect abrupt interface (in which case, all interface Co atoms belong to the monolayer in contact with the Cu layer) to strongly mixed interface regions (which may still contain interface Co atoms with 12 Co neighbors).

To quantify the proportion of hcp- and fcc-like Co in the multilayers as well as the relative intensity of the interfacial part of the spectrum, we fit the spectra to a sum of Gaussian lines, two for the hcp shoulders, one for the fcc main line, and several Gaussians at lower frequencies to take into account the interfacial region, as represented in Fig. 5. The latter lines were constrained to be separated by a gap of 16 MHz as determined for bulk alloys; a different analysis of the interfacial part of the spectrum is presented in the next section, where this gap is freed. From the integrated intensity of the respective Gaussian lines, this first analysis gives us two pieces of information: the proportion of hcp and fcc cobalt in the bulk of the Co layers, on the one hand and, on the other hand, the number of cobalt atoms located in the interface regions of the multilayers. The (15-Å Co)/(20-Å Cu) and (60-Å Co)/(20-Å Cu) multilayers in Table II allow us to compare the proportion of hcp- and fcc-like Co atoms at constant Cu thickness, and the (60-Å Co)/(20-Å Cu) (60-Å Co)/(60-Å Cu), and (60-Å Co)/(90-Å Cu) multilayers at constant Co thickness. The results are summarized in Table II.

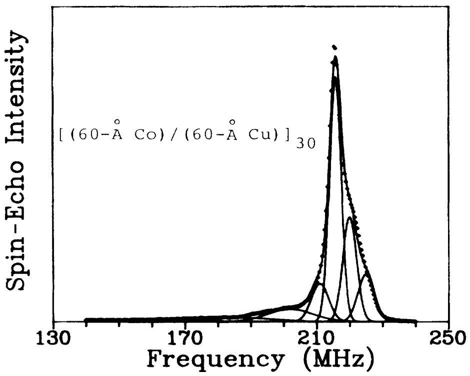

FIG. 5. Example of coarse Gaussian decomposition of a multilayer NMR spectrum.

TABLE II. Proportion of hcp and fcc cobalt environment in the bulk of the layers. For low Co thickness, Co grows with an fcc structure. For large Co thickness, a significant part of Co is hcp. The increase of Cu thickness allows a better recovery of the initial fcc stacking.

Sample	% hcp	% fcc	Fe buffer
(15-Å Co)/(15-Å Cu)	0	100	yes
(15-Å Co)/(20-Å Cu)	0	100	yes
(60-Å Co)/(20-Å Cu)	78	22	no
(60-Å Co)/(60-Å Cu)	44	56	no
(60-Å Co)/(90-Å Cu)	39	61	no

Those results clearly show that an increased Co thickness favors hcp stacking of cobalt while the increase of Cu thickness favors fcc stacking of Cobalt. This observation can be interpreted as follows: At first cobalt grows on top of fcc copper with an fcc structure and then, it gradually changes its stacking into hcp with increasing Co thickness as hcp is the stable Co structure at temperatures below 400 °C. On the other hand, the thicker the copper layers are, the better the fcc stacking of the cobalt layers is. We believe that this last effect is associated with a better reconstruction of the fcc structure of Cu as its layer thickness increases while the initial growth of a thin Cu layer on a badly crystallized thick Co layer is rather perturbed. Since samples with thin Co layers have grown on an iron buffer layer this might also improve the fcc stacking: Other measurements on samples with and without Fe buffer but otherwise identical will be performed to discriminate between the effects of the buffer layer and of the Co thickness.

The proportion of Co nuclei resonating in the low-frequency tail of the spectra gives an approximate knowledge of the quality of the interfaces between cobalt and copper layers, assuming there are no defects in the bulk of the Co layers. Table III gives the number of interfacial cobalt atoms in units of full cobalt monolayers per interface (here after noted FML). The multilayers with thin Co layers show interfaces of 1.7 Co FML while samples with thicker Co layers exhibit more mixed interfaces of about 3.5 Co FML (these values will be discussed in the following section).

In any case, this first analysis of the spectra indicates that the interfaces between Co and Cu are not perfect: The number of interfacial cobalt atoms is larger than 1 FML/interface, which means that the interfaces would be less steep in the present sputtered layers than in previously studied Co/Cu multilayers prepared by UHV eva-

TABLE III. Number of cobalt atoms in the interfaces of the multilayers in units of full Co monolayers per interface (FML).

Sample	Co in the interface (FML)
(15-Å Co)/(15-Å Cu)	1.7
(15-Å Co)/(20-Å Cu)	1.7
(60-Å Co)/(20-Å Cu)	3.0
(60-Å Co)/(60-Å Cu)	3.8
(60-Å Co)/(90-Å Cu)	3.5

poration or molecular beam epitaxy (MBE).[7] Indeed, beside the main line, the NMR spectrum of a multilayer with really flat interfaces would exhibit a single satellite line corresponding to the environment of cobalt atoms at a steep interface (i.e., in case of ⟨111⟩ stacking, nine Co and three Cu nearest neighbors). In addition, this satellite line would arise from only one Co FML. To learn more about the nanostructure of the interfaces, in the following sections we compare the observed spectra with those expected from interface models.

IV. COMPARISON AGAINST INTERFACE MODELS

A. Models

The first interface model we have checked against the experimental spectra is the monoatomic step defect model that we have used for UHV deposited multilayers.[7] This model assumes that the interface roughness only consists in steps which have a monoatomic height. The average distance *d* between these steps and the average length *l* of their straight parts are the two main parameters of the model that describe the in-plane nanostructure of the interface (Fig. 6). The probability of occurrence of the various Co atom neighbor configurations in the interface depend on *d* and *l* according to the expressions given in Table IV.

Secondary parameters of the model are: (1) The frequency gap between lines in the interface spectrum as discussed above for bulk alloys. (2) The highest frequency for Co in the interfaces (Co surrounded by 12 Co). It may be slightly lower than that of Co in the bulk of the layers because of the lower average magnetization in the mixed region (next-nearest- and further-neighbor influence). In this model, it corresponds to a virtual line, as there are no Co atoms with no Cu neighbors in the interface model. This parameter sets the position of the whole interface spectrum with respect to the main line. (3) The width of the supposedly Gaussian lines. (4) And of course the intensity of the whole interface spectrum relative to that of the main line.

Since the coarse analysis has shown that Co/Cu interfaces may be greater than a monolayer, we have also used a general model which introduces a concentration profile through several monolayers. Since it is hard to develop such a model while still taking into account the detailed in-plane structure of the interface, we have assumed that the atomic arrangement of Co and Cu atoms in the mixed atomic layers is random (i.e., in this second model, the interfaces are built from successive two-dimensional random alloys). Each of these interface layers is given a different composition linearly varying from 100% Co to 0% Co through the mixed interface thickness (trapezoidal composition profile). For a given interface concentration profile and following a binomial law, one can

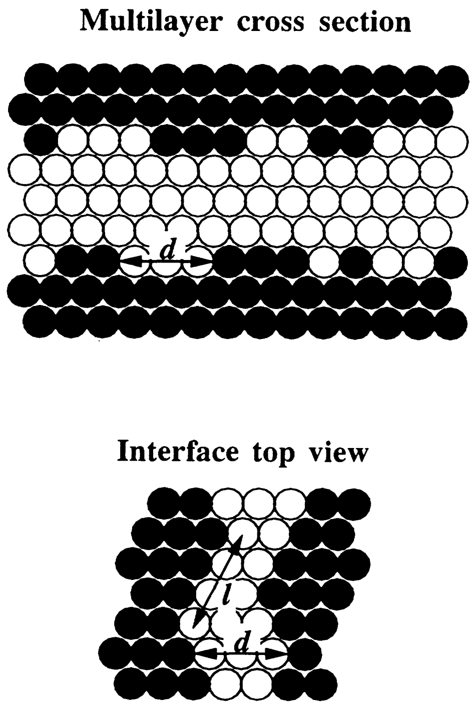

FIG. 6. Schematic view of a multilayer interface with atomic step defects.

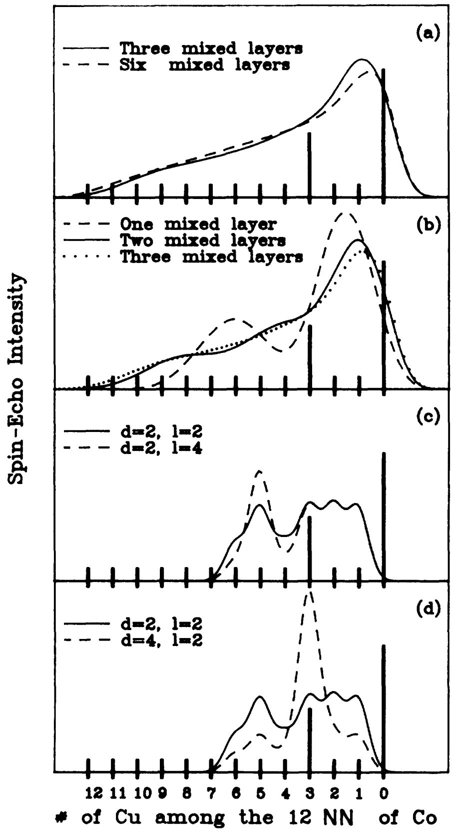

FIG. 7. Expected shapes of the interfacial part of the NMR spectrum for the models tested and various values of the pertinent parameters. (a) and (b) Diffused interface model. (c) and (d) Step interface model. Thick ticks indicate the position of the successive interface lines: their height is arbitrary but it has been enlarged for the mainline (no Cu neighbor) and the perfect interface line (three Cu neighbors).

TABLE IV. Occurrence probabilities of the different near-neighbor coordination of Co for a step interface model and a ⟨111⟩ growth texture; n is the number of Co monolayers per Co layer; d is the average distance between steps; l is the average in plane length of the straight part of the steps.

Cu Neighbors	Intensities Perfect	 Step interface
0	$n/2-1$	$n/2-1-\frac{1}{2d}$
1	0	$\frac{1}{2d}$
2	0	$\frac{1}{2d}$
3	1	$1-\frac{3}{2d}$
4	0	$\frac{1/l}{2d}$
5	0	$\frac{2-2/l}{2d}$
6	0	$\frac{1/l}{2d}$

compute the probability of occurrence of each cobalt nearest-neighbor configuration (from 0 to 12 Cu neighbors); this fixes the relative intensities of all the satellite lines in the interface spectrum and hence, the shape of the low-frequency tail. Thus, such a model has one main adjustable parameter: the slope of the composition profile or accordingly, the number of mixed monolayers within an interface region. Secondary parameters are the same as in the first model. We have tested this last model for six linear concentration profiles, from one mixed layer containing 50% Co and 50% Cu to six mixed layers with Co content ranging from 84% to 17%.

In Fig. 7 are presented the expected shapes of the interfacial part of the NMR spectrum for the models tested and various values of the pertinent parameters. In these simulations we have voluntarily used rather large individual linewidths in order to put the stress on differences between overall spectrum shapes expected from the models. It can be seen that these shapes are quite different between the step interface model and the diffused models. Among the diffused models differences can be observed up to an interface thickness of three monolayers. For three mixed layers and above one cannot discriminate at day light between the models; then one can rely only on fit-agreement factors and on checks based on the relative intensity of the interfacial tail compared with that of the full spectrum (coarse analysis).

B. Application

Following the indications of the coarse analysis, the second model was first applied to the multilayers with low cobalt thicknesses [(15-Å Co)/(20-Å Cu), (20-Å Co)/(20-Å Cu)], where the interface contribution to the spectrum is relatively more important. The fits for those multilayers are satisfactory with an interface thickness parameter of three mixed monolayers, less satisfactory for larger thicknesses, and completely unsatisfactory for a one-monolayer-thick interface, as shown by the reconstructed spectrum of the (15-Å Co)/(15-Å Cu) multilayer in Fig. 8 and by the χ^2 values in Table V. The model spectra for (15-Å Co)/(20-Å Cu) multilayer gave the same results. For comparison with the coarse analysis the number of Co in the corresponding thick interfaces is given in Table V. It can be seen that the results of the two analyses are consistent, as the coarse analysis yielded a number of cobalt in the interfaces of 1.7 Co FML while the model with three mixed layers corresponds of 1.9 Co FML contributing to the interface spectrum.

In all instances (samples and models) the best fits yield a gap of about 15 MHz between each line of the interface. This value is much closer to that found in Co-Cu alloys than in epitaxial Co/Cu superlattices,[7] which is certainly a consequence of the absence of well-defined growth texture and possibly of stronger Co-Cu admixture in the sputtered layers.

Even though the three-monolayers-thick interface model reproduces the overall spectrum shape well, in Fig. 8(b) one can see that the fourth line intensity of the model (three Cu neighbors) is too weak compared to the adjacent lines to fit the spectrum really correctly. It means that there are more Co atoms in an environment comprising nine Co and three Cu nearest neighbors than the number anticipated from our model. This environment corresponds to interfacial cobalt in a flat, fcc ⟨111⟩ oriented interface. So, it seems that our samples have lo-

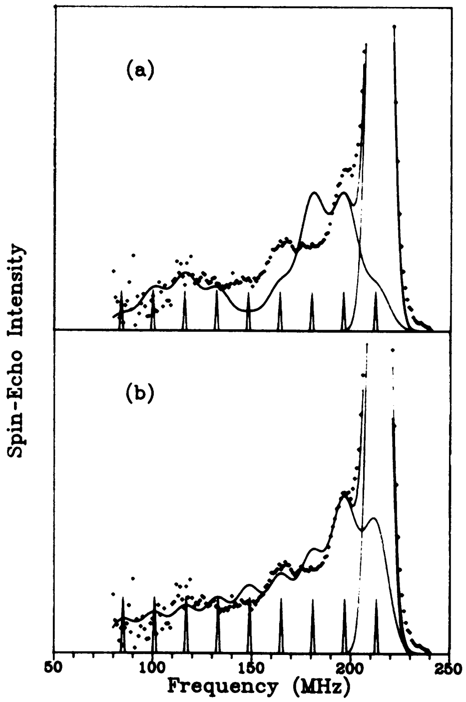

FIG. 8. Modeling of the interfaces of the (15-Å Co)/(15-Å Cu) multilayer with the diffused interface model. (a) Example of fit with an interface thickness of one monolayer. The best fit is obtained with an interface thickness of three monolayers (b). Triangular ticks indicate the position of the successive interface lines.

TABLE V. Number of Co atoms in the interface calculated for the diffused interface model in units of full Co monolayers per interface (FML); and χ^2 values obtained with this model applied to the (15-Å Co)/(15-Å Cu) multilayer. The best fit (i.e., the smallest χ^2) is obtained for an interface thickness of three layers.

Thickness of the interface (monolayers)	Number of Co in the interface (FLM)	(15-Å Co)/(15-Å Cu) χ^2
1	1.3	11.8
2	1.6	3.9
3	1.9	2.3
4	2.2	2.5
5	2.6	3.1
6	3.0	4

cally flatter interfaces than those allowed by the model of random in-plane chemical short-range order. We believe that an extension of the monoatomic step interface model to a model of mesalike islands of Cu (or Co) with distributed height (instead of monoatomic thick rafts) can account for the excess intensity of the three Cu neighbor line. Such nanostructure would be also more consistent with the Co-Cu repulsive interaction than the solid solutionlike structure tested here. Yet, we prefer another possible explanation, which is suggested by the analysis of the results obtained for samples with thick Co layers and which is given in the following paragraphs.

The spectra observed for samples with thicker Co layers can hardly be described by any interface model we have imagined so far. Indeed, the significant shoulder observed on the low-frequency side of the mainline suggests that there is a contribution of Co atoms with an environment of 11 Co nearest neighbors larger than allowed by the models. Even if we use an interface concentration profile extending over six mixed layers (3.0 FML) which gives the best agreement with the experiment, the fit is still obviously unsatisfactory [Fig. 9(a) and Table VI]. The observed extra intensity of the shoulder seems to be typical of sputtered samples with thick Co layers, as we have also observed in sputtered Cu/Co/Cu sandwiches from another source. The only distribution of Co and Cu atoms in the interface which would give such a large contribution of Co nuclei with one Cu neighbor is a two-dimensional ordered compound Co_2Cu. In such a case all Co atoms in the mixed layer would have six Cu neighbors (three in their plane and three in the adjacent pure Cu layer) and all Co atoms in the adjacent pure Co monolayer would have one Cu neighbor: Such an extreme situation would correspond to about 60% of the interfacial spectrum intensity lying in the one neighbor line but it seems totally unrealistic.

We have consequently tried to model the spectra using the same interface models along with a separate additional line to take the shoulder into account. Good fits were then obtained for an interface thickness of one randomly mixed layer (i.e., 1.3 FML in the interface) and the additional line containing 1.5 FML [Fig. 9(b)]. The 3.5 FML found by the coarse analysis can then no longer be attributed only to the interface, unless the additional line also arises from the interface region for which we have neither evidence nor explanation.

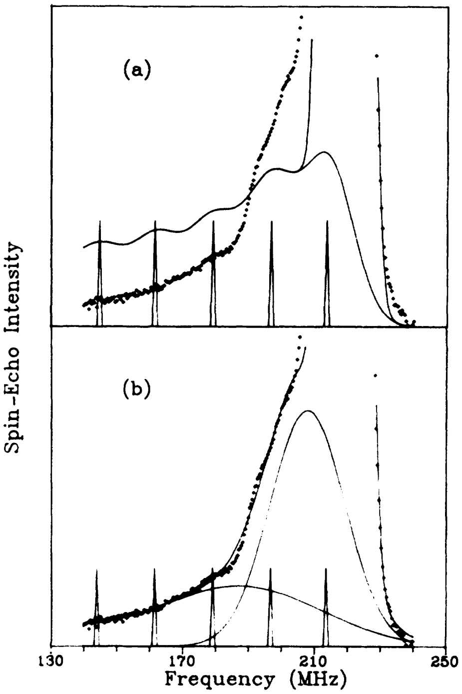

FIG. 9. Modeling of the interfaces of the (60-Å Co)/(90-Å Cu) multilayer. The model of diffused interfaces (a) does not fit the experimental data. We have to add an extra impurity line (b) corresponding to Co with an environment of 11 Co nearest neighbors to reproduce correctly the spectrum.

In the light of the results obtained for thick Co layers and the possible presence of bulk defects, the spectra observed in samples with thin Co layers have been reconsidered and an additional line at around 200 MHz was allowed in the models. With the help of such supplementary degree of freedom, an excellent fit (Fig. 10 and χ^2 value, Table VI) was obtained with a step interface model close to the one suggested for UHV-grown layers ($d = 2$

TABLE VI. Best χ^2 values for the different interface models and for the two types of multilayers. The table gives: value of χ^2 (upper figure) and number of mixed monolayers in the interface (lower figure). Model 1: diffused interface model. Model 2: single diffused layer with additional line. Model 3: step interface with additional line.

χ^2 Interface	Model 1	Model 2	Model 3
(15-Å Co)/(15-Å Cu)	2.3 3 ML	1.8 1 ML	1.6 1 ML
(60-Å Co)/(90-Å Cu)	160 6 ML	1.8 1 ML	

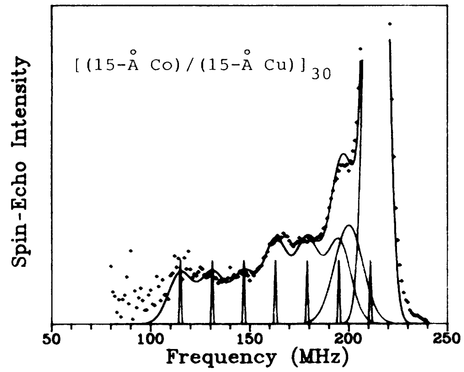

FIG. 10. Modeling of the interfaces of the (15-Å Co)/(15-Å Cu) multilayer by a step interface model and an additional line corresponding to Co with an environment of 11 Co nearest neighbors in the bulk of the Co layers.

atomic distances, $l = 1.5$ atomic distances), provided that the additional line is accounted for by defects in the bulk of the layers that affects 0.2 FML Co. The step interface model was not considered for samples with thicker Co layers because the hcp-fcc stacking admixture results in broader satellite lines and its does not allow a reliable determination of d and l.

For the five samples we have studied, the concentration of Co nuclei resonating in the extra line amounts to about 9%, which clearly suggests a bulk nature of the defects responsible for the occurrence of this line. Of course, a continuous range of Co thicknesses would be needed to ascertain a scaling between the thickness and the intensity of the extra line in order to confirm the bulk origin of this line. The origin and nature of such bulk defects could be of two kinds. Firstly, there may be diluted impurity atoms in the bulk of the Co layers such as copper or argon atoms or even vacancies; we have no reference for the influence of an argon or a vacancy neighbor on the Co hyperfine field, but it is reasonable to assume that their effect is close to that of Cu (nonmagnetic, non-*d* element). Considering that one point defect in the bulk affects the resonance frequency of 12 Co nuclei, the concentration of nonmagnetic impurities, which can be deduced from the intensity of the additional line, is about 0.7% in all samples. Secondly, Co nuclei at grain boundaries may also experience a lower hyperfine field than in the grains. Considering that the structure of sputtered multilayers is generally more coherent along the growth direction than in the layer plane (columnar structure) we can, from the ratio of the number of surface atoms to the number of volume atoms in a column, deduce that such columns would have an average diameter of about 90 Å, which is a quite reasonable figure.

V. CONCLUSION

Using zero-field NMR we have analyzed the in-layer structure and the interface quality of cobalt-based multilayers, which provides additional local information on atomic distributions in buried layers and complements the information obtainable by other structural investigations.

Through this study, the role of the thickness of each component on the structure of the multilayers is demonstrated. Only multilayers with low cobalt thicknesses show purely fcc cobalt stacking and with increased Co thickness, the more stable hcp phase starts to build up. The role of Cu in stabilizing the fcc stacking of the multilayer is also evidenced as the proportion of hcp Co decreases with increasing Cu thickness. Complementary experiments are needed to investigate the role of an Fe buffer layer.

The nanostructure of the interfaces between Co and Cu layers has also been modeled and can be described as follows. If the presence of defects in the bulk of the Co layers is excluded, the NMR spectra observed in samples with thin Co layers must be explained by an intermixing of Co and Cu over three monolayers at the Co/Cu interfaces. However, such a model does not hold true for samples with thick Co layers where the existence of bulk defects is strongly suggested. In this case, the spectra for all the studied samples can be explained by sharp Co/Cu interfaces (one mixed layer) and about 0.7% point defects or impurities (Ar?) in the bulk of the layers or possibly columnar grains with an in-plane diameter of about 90 Å. The role of bulk defects or impurities on the magnetoresistive properties of Co/Cu multilayers (and other sandwiches) has been suggested by some authors;[12] it would be interesting to check if there is a quantitative correlation between the magnetoresistance of various systems and the number of defects determined by the present method.

ACKNOWLEDGMENTS

We wish to acknowledge useful comments from Dr. P. A. Schroeder and Dr. W. P. Pratt, Jr. The sample preparation was funded in part by the Center for Fundamental Materials Research of Michigan State University and the U.S. NSF under Grant No. DMR-88-13287.

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