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Magnetic layer thickness dependence of the interlayer exchange coupling in (001) Co/Cu/Co

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A dependence of the strength of the antiferromagnetic coupling across Cu on the *Co layer thickness* has been observed. The Co thickness dependence displays two clear peaks consistent with the recently predicted oscillation period of 6.2 Å Co. Apart from the two peaks also several small peaks are visible on a scale of about 1 monolayer Co. Free-electron calculations indicate that these rapid variations in strength may result from slight differences between the slopes and starting points of the two Co wedges that were involved in the experiment.

Recent theoretical work by $Bruno^1$ and $Barnas^2$ has shown that the interlayer exchange coupling between two ferromagnetic (FM) layers across a nonmagnetic (NM) metallic spacer layer may oscillate not only with the thickness of this spacer but also with the thickness of the ferromagnetic layers.

Systematic experimental studies investigating the detailed effect of the FM thickness upon the interlayer coupling are very scarce. Qiu et al.³ and Chen et al.⁴ studied the Co/ Cu/Co (100) system for several Co thicknesses. However, their results were insufficient to reveal an oscillatory behavior. First experimental evidence for an oscillatory behavior as a function of the magnetic layer thickness was obtained by the present groups for a (001) Co/Ni/Co/Cu/Co/Ni/Co sandwich⁵ and by Okuno and Inomata for Fe/Cr (100) multilayers.⁶ In this article we summarize our experimental results⁵ and discuss them using calculations based on the Bruno model.¹ Within the free-electron approximation, this model could be extended to include our experimental situation viz. a situation of, in principle, unequal FM layers which, in addition consist of multiple different FM layers (three in our case, Co/Ni/Co). To interpret or predict the behavior of these at first sight more complicated systems it is useful to recall the mechanism from which an oscillation with a FM layer thickness originates.

Bruno shows that the coupling problem can be described in terms of the reflection of electron waves at the potential steps at the various interfaces in the FM/NM/FM sandwich.⁷ Here, the nonzero exchange spitting of the conduction bands in the FM layers is responsible for a difference in potential step heights for spin-up and spin-down electrons. This causes the reflection amplitude to be spin dependent resulting in a magnetic coupling. The coupling strength is larger for larger differences between the reflection amplitudes for spin-up and spin-down electrons. An oscillatory dependence of the coupling strength on the thickness of the FM layers is then, as Bruno argues, simply a result of multiple reflections of electron waves within the FM layers.¹ As in the case of light waves incident on a (multi)layer, the effective reflection amplitude of a layer (in our case a FM layer) is a result of the constructive and destructive interferences of the forward (transmitted) and backwards (reflected) traveling waves and depends upon the interplay between the layer thickness, the wavelength of the incident wave, and the wavelength within the reflecting medium. The latter is determined by the electronic structure of the FM layers and follows, in the large thickness limit, from the relevant extremal Fermi surface (FS) spanning vectors of the FM layer. From this "electronoptics" picture it is thus clear that if a FM layer is composed of, for example, a multilayer made of several different FM layers, the effective reflection amplitude of such a multilayer (and thus the coupling) will oscillate with the thickness of any of the constituent FM layers. This is exactly the case for our experimental system.

We have studied a molecular-beam-epitaxy-grown (001) Co/Ni/Co/Cu/Co/Ni/Co sandwich in which the two Co layers adjacent to the Cu spacer as well as the Cu spacer itself were deposited in the form of wedges oriented perpendicularly with respect to each other. This allowed for independent investigation of the Cu and Co layer thickness dependence of the coupling across Cu(001) in a single sample. In this way experimental artifacts are avoided that are related to changes in (i) deposition conditions, (ii) substrate quality, and (iii) layer thickness-changes that otherwise would have occurred in a series of separate samples. Especially slight changes in a presumably fixed Cu spacer layer thickness in an experiment of varying magnetic layer thickness would cause problems in our case since the coupling strength is extremely sensitive to the precise Cu thickness because of the presence of a short period oscillation with a period of about 2.6 ML Cu. Note that the latter problem was not encountered in the study of the sputtered Fe/Cr (100) samples of Okuno and Inomata⁶ since only the long Cr period was present in their samples.

Two samples (referred hereafter as samples I and II) have been investigated. The typical composition of the magnetic layers in the samples was as follows: 30 Å Co/15 Å Ni/Co wedge (2.3 Å/mm). Further compositional details and information regarding the structure of the layers as obtained from low energy electron diffraction experiments can be found in Ref. 5.

The antiferromagnetic (AF) coupling behavior was in-



FIG. 1. The strength of the interlayer exchange coupling in the first and second AF peak as a function of the Co thickness for sample II.

vestigated by measuring hysteresis loops via the longitudinal magneto-optical Kerr effect. The behavior as a function of the Cu thickness displays a superposition of a long and a short period oscillation,⁵ in accordance with earlier observations.^{8,9} The presence of the short period in both samples indicates that these samples are of high structural quality. The dependence on the Co thickness of the strength of the first two AF maxima (at ≈ 9 Å Cu and ≈ 19 Å Cu) for sample II is shown in Fig. 1. An oscillatorylike behavior with an apparent period of 6-7 Å is observed in all experimental scans. From the electron-optics picture of Bruno it is immediately clear that this value is a property solely of Co. With varying Co thickness the effective reflection amplitude of the Co/Ni/Co FM layer (and thus the coupling) is modified in an oscillatory fashion with a period determined by the relevant wavelength in the Co layer. According to Bruno the latter is determined by the extremal spanning vector along the Γ -X line [(100) growth] in the spin-down FS of fcc Co. This vector yields a period of 3.5 ML or 6.2 Å Co which is in very good agreement with the present experiment. However, the functional shape of the experimentally obtained variation with Co thickness does not resemble a fully regular oscillatory behavior. Considering for example the behavior of the second AF peak, the lower curve in Fig. 1, two clear peaks are visible whereas around the position where the third peak is expected three smaller peaks occur with a spacing of about 2 Å. This behavior seems in contradiction with theory. In particular from the aliasing effect one would expect that because of the sampling at discrete Co planes, periodic variations in coupling strength should only occur on a scale larger than 2 ML (3.6 Å). This is true in the ideal situation. However, in the present experiment two Co wedges are involved which may not be identical but may differ slightly in, e.g., their slope. In this respect we remark that with scanning Auger electron spectroscopy (AES) the slopes were determined to be equal within 10% accuracy. In order to evaluate the effect of unequal Co wedges on the experimentally observed behavior and in particular if it is possible to explain variations in coupling strength on a scale of 1 ML Co, we have extended the model of Bruno¹ to our experimental geometry and performed a number of simulations. Before proceeding with the results we briefly describe the steps that we have undertaken to adapt the model to our situation.

First, to account for unequal wedges Eq. (4) in Ref. 1 is generalized to the case of unequal FM lavers. As is clear from Ref. 1 this can be performed by a Taylor expansion of the first Eq. in Ref. 1. In principle this step is sufficient to evaluate if rapid variations in coupling strength may arise from unequal wedges. Second, to account for the effect of the two additional FM layers which are coupled to the Co wedge the model is extended to the case that each FM layer is composed of an arbitrary number of layers, i.e., to describe the system $FM_1^a/FM_2^a/\dots/FM_k^a/spacer/FM_1^b/FM_2^b/\dots/FM_1^b$ with k and l integers denoting the number of FM layers of which FM layers a and b are composed, respectively. To our knowledge the latter step can only be made easily within the free-electron approximation. Within this approximation it is straightforward to calculate the effective reflection amplitude of each FM multilayer. Continuity of the wave functions (plane waves) and their derivative at the interfaces directly enables one to write down a recursion relation for the reflection amplitude of an arbitrary multilayer. Using this relation instead of the Fabry-Pérot formula given by Bruno [Eq. (5) in Ref. 1] allowed us to calculate the Co thickness dependence of the coupling for the situation in which the slopes of the Co wedges differ and their starting point do not coincide.

The results of several calculations for 19.86 Å Cu, i.e., for the strength of the second AF peak, are shown in Figs. 2(a)-2(c). The calculated coupling strengths are normalized to the limit of infinite Co wedge thickness. In Fig. 2(a) the behavior is shown for the ideal case of two identical Co wedges. Here the aliasing effect is demonstrated for the Co dependence. The fundamental period $\lambda = \pi/k_{F}^{2} = 2.49$ Å pertaining to the Fermi wave vector k_{\pm}^{\downarrow} of the spin-down fcc Co FS, yields after aliasing (with 1.805 Å Co ML thickness) a period of 6.1 Å. In Fig. 2(b) the ideal case is again calculated. However, for this calculation the situation that a Co layer consists of a nonintegral number of monolayers (incomplete coverage) is also calculated. This is done from a linear combination of the coupling across two independently patchy interfaces. Incomplete coverage is thus treated as follows: We define J(n,m) as the coupling strength for the combination of n integral number of Co monolayers in Co wedge A and m monolayers in Co wedge B. The coupling $J'(t_A, t_B)$ for the situation that the thicknesses t_A and t_B at Co wedge A and B, respectively, are a nonintegral number of monolayers is calculated from

$$J'(t_{A},t_{B}) = J(n,m)(1-f_{A})(1-f_{B}) + J(n,m+1)$$

×(1-f_{A})f_{B}+J(n+1,m)
×f_{A}(1-f_{B}) + J(n+1,m+1)f_{A}f_{B}.

Here, $f_{A(B)}$ represent the fractional coverages defined by $t_A = n + f_A$ and $t_B = m + f_B$ with $0 \le f_{A(B)} \le 1$. Considering Fig. 2(b) it is clear that even for the ideal case of equal wedges, fractional coverages result in additional peaks like the small one between the third and fourth monolayer (± 6 Å). Such features are a direct result of the asymmetric Co layer thick-

7082 J. Appl. Phys., Vol. 76, No. 10, 15 November 1994

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FIG. 2. Free-electron calculations of the Co thickness dependence of the coupling strength for 30 Å Co/15 Å Ni/Co/19.86 Å Cu/Co/15 Å Ni/30 Å Co. The values are normalized to the strength of infinite Co thickness limit. Calculation parameters are k_{c}^{α} =1.471 Å⁻¹, k_{f}^{Lo} =1.261 Å⁻¹, k_{f}^{fCo} =1.363 Å⁻¹, k_{f}^{IN} =1.362 Å⁻¹, k_{I}^{IN} =1.389 Å⁻¹ (Ref. 10).

ness combinations [(n, n+1) and (n+1, n)]. It thus appears that for the present case in which two active layers are involved one is not allowed to make a linear interpolation [such as in Fig. 2(a)] between the situation in which both Co layers are n ML thick (n,n) and the situation in which both Co layers are n+1 ML thick [(n+1, n+1)]. The experimentally observed rapid variations in coupling strength may originate from this phenomenon. In an attempt to fit a freeelectron calculation to the experimental strength dependence of the second AF peak (Fig. 1) we have tried a number of combinations for the wedge slopes and starting points of the Co wedges. Figure 2(c) shows a calculation in which the Co wedge slopes differed by about 10% from the AES determined value of 2.3 Å/mm viz. 2.5 Å/mm for Co wedge A and 2.1 Å/mm for Co wedge B. In addition wedge A has been given an offset of 0.5 Å with respect to wedge B. From the combinations we have tried it appeared that the first two peaks are relatively insensitive to modifications of the wedge parameters so that their separation remains a good measure for the oscillation period. The behavior at the larger Co thicknesses appears more susceptible. From Fig. 2(c) it is clear that the third and fourth peak may even disappear. Instead three smaller peaks appear with a spacing which is considerably smaller than the 6.2 Å oscillation period. The qualitative agreement with the experiment is striking, suggesting that the originally proposed interference/beating ef-



FIG. 3. Free-electron calculation of the Co thickness dependence of the coupling strength for the first AF peak, i.e., for 30 Å Co/15 Å Ni/Co/7.22 Å Cu/Co/15 Å Ni/30 Å Co. The values are normalized to the strength of infinite Co thickness limit.

fects in Ref. 5 may not be necessary to explain the behavior at the larger Co thicknesses. We did not attempt to obtain a better fit by varying more parameters, e.g., by introducing a difference in the thicknesses of the two Ni layers which were adjacent to the Co wedges. One should realize that the present model, although it explains many of the observed features, is a free-electron approximation and therefore seems inappropriate to make a comparison with the experiment on a detailed level. This is also reflected by the calculation shown in Fig. 3 representing the behavior for the first AF peak (calculation at 7.22 Å Cu). Here, the same Co wedge parameters were used as those to obtain the reasonable fit for the second AF peak [Fig. 2(c)]. The simulation does not display the smaller sharp features at the higher Co thicknesses such as observed in the experiment [the upper curve in Fig. 1]. Instead the variations are more gradual. What does agree are again the important features. Apart from the ones we already mentioned it is seen that the relative oscillation amplitude for the first AF peak is smaller than that for the second AF peak, (compare Figs. 3 and 2(c)). This is in agreement with the experiment where the peaks as afunction of the Co thickness are more pronounced for the second AF peak than for the first AF peak (Fig. 1).

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- ¹⁰ We obtained these Fermi wave vectors from self-consistent ASW bandstructure calculations.

Bloemen et al. 7083

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