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Biperiodic oscillatory coupling as a function of the thickness of an embedded Ni layer in Co/Cu/Co/Ni/Co(100) and selection rules for the periods

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A biperiodic oscillation of the strength of the antiferromagnetic interlayer coupling as a function of the thickness of an embedded Ni layer has been observed in an epitaxial Cu(100)/Co/Cu/Co/Ni/Co sample with the Cu interlayer and the Ni layer in the form of wedges. As the effect originates from Bloch-wave interference in the Ni layer, the observed periods must be, and indeed can be, related to extremal spanning vectors of the spin-resolved Ni Fermi surface. The experiment touches on the selection criteria for spanning vectors of Ni that determine the periods of the oscillations. [S0163-1829(96)50726-5]

Many aspects of the oscillation of the interlayer coupling as a function of the spacer thickness in magnetic multilayers are well understood by now.[1] Initially, the period was shown to be determined by extremal spanning vectors (callipers) of the spacer Fermi surface (FS). Recently, the dependence on the thickness of the ferromagnetic[2–4] and cap layers[5–7] has been investigated. These experiments confirmed existing theories that are based on spin-dependent reflection of electron waves in the whole multilayer stack.[8,9] Further, they also raised the question as to which of the callipers of the FS of the ferromagnet or cap layer material determines the period. For example, the minority spin FS of fcc Co alone has ten different callipers along the (100) direction,[2] which are all potential candidates for defining oscillation periods. Currently, selection rules receive explicit attention[10] and state that the in-plane wave vector $k_\parallel$ must be conserved for callipers of the spacer FS and the FS of the ferromagnet or cap layer material.[11] So far, all experiments automatically obeyed this selection rule, due to the fact that at $k_\parallel = 0$ the FS's usually have callipers[2–6] or because identical spacer and cap layer materials ensure a perfect match of the FS's.[7] In the case of Cu and Ni FS's also a perfect match exists for the callipers at $k_\parallel = 0$ (responsible for a long period oscillation), but for the callipers at $k_\parallel \neq 0$ (short period) the $k_\parallel$ probably differ. Therefore, a study of the dependence of the coupling across a Cu interlayer as a function of a ferromagnetic Ni layer may or may not reveal a short period oscillation and shed some light on the underlying selection rules.

In this paper we report on an experiment that challenges the selection rules. A long and short period oscillation in the interlayer coupling as a function of the thickness of a Ni layer have been observed. As the calliper of the FS of Ni and Cu probably do not share the same $k_\parallel$ this means that $k_\parallel$ is not conserved. To explain this deviation from theory several suggestions are made. Moreover, the experiment extends the study of the dependence of the coupling on the ferromagnetic layer thickness for the magnetic transition metals,[2–4] and confirms that layers that are not adjacent to the spacer also contribute to the coupling strength.

To measure the dependence of the coupling on the Ni thickness, the following multilayer was deposited by molecular beam epitaxy (MBE) on a Cu(100) single-crystal:

$$\mathrm{Cu}(100)/40\ \text{Å}\ \mathrm{Co}/\mathrm{Cu}\ \text{wedge}(0-35\ \text{Å})/4\ \text{Å}\ \mathrm{Co}/$$

$$\mathrm{Ni}\ \text{wedge}(0-25\ \text{Å})/30\ \text{Å}\ \mathrm{Co}/10\ \text{Å}\ \mathrm{Cu}/20\ \text{Å}\ \mathrm{Au}.$$

A schematic picture of a very similar structure is given in Ref. 2. We have deliberately *not* used a sample with symmetric magnetic layers, i.e., with two Ni wedges, because the inevitable inequality of the wedge slopes and starting points can obscure the oscillatory behavior.[12] A thin Co layer was inserted between the Cu and Ni wedges as the exchange coupling in Co/Cu/Co is larger than in Co/Cu/Ni. In addition, on top of the Ni wedge a 30 Å Co layer was deposited to be able to align the coupled magnetic moments on both sides of the Cu interlayer at small Ni thickness. Both added Co layers facilitate the measurement and interpretation of the Kerr hysteresis loops from which the coupling strength is determined. To allow independent variation of the Cu and Ni thickness the wedges were arranged perpendicularly. Growth conditions and the sample analysis are the same as in Ref. 2. The

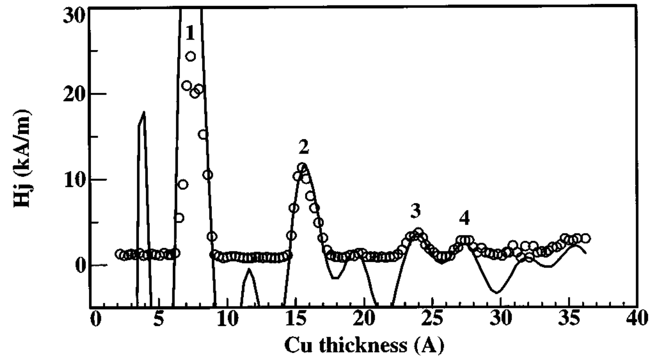

FIG. 1. Dependence of the coupling field H_J on the thickness of the Cu interlayer at zero Ni thickness. The solid line is a fit based on the phenomenological equation (1).

analysis yielded a Cu wedge slope of 4.2 ± 0.6 Å/mm and a Ni wedge slope of 3.2 ± 0.5 Å/mm.

First the dependence of the coupling strength on the interlayer thickness was studied at different Ni layer thicknesses to locate antiferromagnetic (AF) coupling peaks and to trace their position with changing thickness of the Ni layer. The result of the scan before the Ni wedge is shown in Fig. 1. Here the coupling field H_J, defined as the field where the Kerr effect reaches half its saturation value, is plotted against the thickness of the interlayer. Clearly, a long and short period oscillation are present, as may be judged from the peak width relative to the peak separation (peaks labeled 1, 2, and 3), the separation of peaks 3 and 4, signs of peaks between 2 and 3 and beyond 4, and finally from comparison with other experiments.[13]

To obtain the values of both periods the data were fitted with the following, phenomenological equation:

$$J=J_o+\sum_{i=1,2}\frac{J_{o,i}}{(t+t_o)^2}\cos\left(\frac{2\pi t}{\Lambda_i}+\phi_i\right), \qquad (1)$$

where $i=1,2$ refers to the long and short period. For comparison with the experiment a conversion of J to H_J is included in the fitting procedure, incorporating the effect of unequal magnetic moments per area $t\mu_o M_s$ on either side of the Cu spacer. For relatively high anisotropy compared to the coupling (second AF peak or higher), $H_J = J/t\mu_o M_s$ with $t\mu_o M_s$ the smaller magnetic moment per area, was used. For the first AF peak, relatively low anisotropy, H_J was calculated from Eqs. (7) and (8) of Bloemen *et al.*[14] For different combinations of the phases (ϕ_i) and periods (Λ_i), the amplitudes ($J_{o,i}$) were fitted in order to achieve a minimum deviation from the positive experimental values (negative values of the fit are not considered). Only positive H_J or AF coupling can be measured in the present case of a sample of effectively two magnetic layers.

In the fit of the interlayer dependence the Cu thickness (t_{Cu}) was substituted for t and $J_o=0$ and $t_o=0$ were used. A correct fit of all peak heights appeared to be impossible on the basis of Eq. (1) and the best fit (solid line in Fig. 1) was obtained by overestimating the height of the first peak. The lower experimental value may be a result of averaging of the coupling strengths at different thicknesses due to roughness, or of ferromagnetic bridges (pinholes) at small Cu thicknesses. Nevertheless, the peak positions of the fit agree very well with those of the experimental peaks. The values for the periods were 9.0 ± 0.5 Å and 4.0 ± 0.2 Å, excluding a 15% uncertainty in the wedge slope. Comparison with the theoretical periods of 10.6 Å (corresponding to the calliper at the ‘‘belly’’) and 4.7 Å (at the ‘‘neck’’),[15] shows that the ratio of the periods is correct (both 0.44) but the absolute values are too low. This indicates that the actual wedge slope is probably somewhat larger than the measured one, but still lies within the limits set by the experimental accuracy. For the following, however, it is sufficient to note that two oscillation periods are observed.

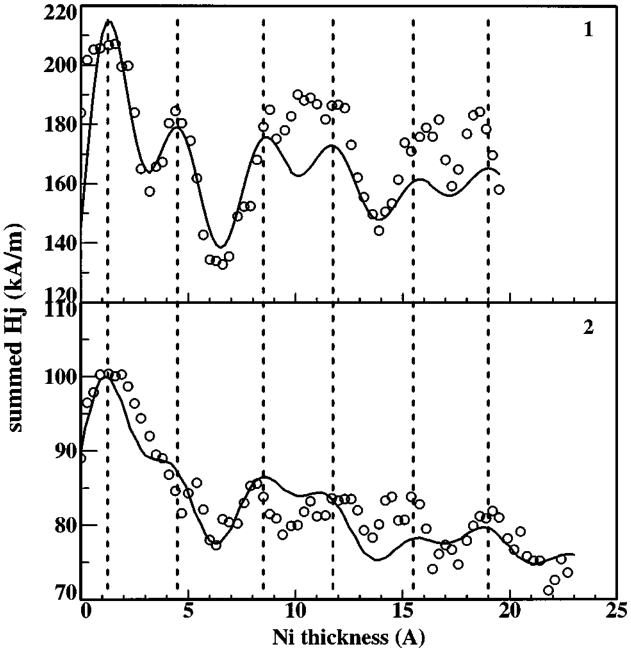

FIG. 2. Dependence of the summed coupling fields H_J on the thickness of the Ni layer at the Cu peaks labeled 1 and 2 in Fig. 1. The solid lines are fits based on the phenomenological equation (1) and the dashed lines indicate the maxima of the oscillations.

At interlayer thicknesses corresponding to the AF coupling peaks labeled 1 and 2 in Fig. 1, Kerr loops were measured as a function of the thickness of the Ni layer. To be sure to find the maximum AF coupling at each Ni thickness, a series of 9 Kerr loops was measured as a function of the Cu thickness across each AF coupling peak at positions separated by 0.05 mm (0.2 Å Cu), thus approximately spanning the full width at half maximum of the AF peaks in Fig. 1. However, in order to reduce noise the sum of the coupling fields obtained from the 9 loops is considered. In Fig. 2 the summed AF coupling fields are plotted as a function of the Ni layer thickness at the first two interlayer AF maxima. A significant variation is seen, which cannot be characterized by a single, long period.

On theoretical grounds a double period oscillation, originating from the interference of electron waves in the Ni layer, is expected, based on two callipers one at the ‘‘neck’’ and one at the ‘‘belly’’ of the Ni FS, as for Cu. Therefore, a fit with Eq. (1), but in this case with the Ni thickness substituted for t, is attempted. An offset coupling J_o is also fitted now and as a first approximation we take $t_o = t_{\mathrm{Cu}} + 4$ Å Co.

TABLE I. Callipers derived from calculated Ni FS's and the ASW calculation performed by the authors. The half of the calliper $k_\perp$ as a fraction of ΓX, the corresponding aliased period Λ, the corresponding in-plane wave vector $k_\parallel$ as a fraction of ΓK, and the spin are given. The uncertainty in the values for the long and short periods as a result of measuring the lengths of callipers are estimated as 0.5 Å and 0.1 Å, respectively.

$k_\perp$ (ΓX)	Λ (Å)	$k_\parallel$ (ΓK)	Spin	Author(s)
0.76	7.4	0	Down	
0.74	6.7	0	Up	Connolly (Ref. 16)
0.49	3.6	0.56	Up	
0.80	8.9	0	Down	
0.76	7.5	0	Up	Tsui (Ref. 17)
0.50	3.6	0.50	Up	
0.75	7.0	0	Down	Callaway
0.79	8.3	0	Up	and Wang (Ref. 18)
0.51	3.6	0.57	Up	
0.75	7.5	0	Down	
0.77	8.0	0	Up	ASW calculation
0.52	3.7	0.52[a]	Up	fcc Ni
0.52	3.7	0.54	Up	

[a] $k_\parallel$ corresponding to the calliper of the short period in Cu.

This choice of t_o and the functional dependence on t_o are motivated in a study of the cap layer.[5] Also here, the conversion from J to H_J is implemented. Due to the varying thickness of the Ni layer this results in a slightly decreasing background with increasing Ni thickness, as is visible in the fits. The best fits on the basis of (1) are shown as solid lines in Fig. 2.

It appeared that the behavior at both AF peaks could be described with one set of oscillation periods (3.6 ± 0.2 Å and 7.9 ± 0.5 Å) and phases. In the error margins the uncertainty in the Ni wedge slope (15%) is not included. This correlation of periods and phases is indicated by the dashed lines through the maxima. As the periods are all determined by the Ni FS they must be the same, of course. The fact that the phases of the oscillations appear not to depend on the Cu thickness, can be explained along similar lines as in a recent cap layer study.[5] The variable part of the phase is determined by the Cu spacer thickness. Between peak 1 and 2 in Fig. 1 this phase difference is approximately a multiple of 2π for both the short and long period. Therefore, there appears to be no phase shift at all between oscillations 1 and 2 in Fig. 2. Again, the fits do not match the data in the whole thickness range, perhaps a consequence of the summing procedure in this case, but the positions of the maxima and minima are reproduced well. As the periods are our main concern, we will leave the discussion of the quality of the fits in relation to possible imperfections of Eq. (1) for now.

To relate the experimentally observed periods to callipers of the spin-up or spin-down FS of Ni, we have summarized the relevant values in Table I. We have also carried out self-consistent augmented spherical wave (ASW) calculations in the local density approximation. These results are also listed in Table I. From the calculated Ni FS's the callipers and the corresponding aliased periods could be derived by measuring their length along ΓX. For clarity in Fig. 3 a schematic representation of the relevant cross section (''dogbone'') of the Cu and spin-up Ni FS is given. The long period ranges from 6.7 Å to 8.3 Å for spin-up electrons and from 7.4 Å to 8.9 Å for spin-down electrons. Much better agreement is found between calculations of the calliper at the ''neck'' that is responsible for a short period of 3.6 to 3.7 Å. However, the corresponding in-plane wave vector $k_\parallel$ of the short period ranges from $0.50\Gamma K$ to $0.57\Gamma K$. No calliper giving rise to a short period exists for spin-down electrons. Due to aliasing, the long period is much more sensitive to the position of the Fermi level than the short period, leading to a larger uncertainty in the long period.

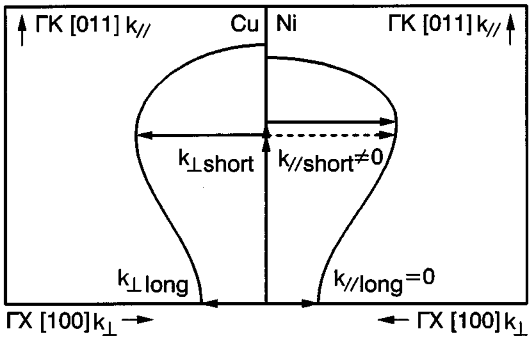

FIG. 3. Schematic representation of the relevant cross section (''dogbone'') of the Cu and spin-up Ni FS. The long and short period callipers $k_\perp$ along ΓX, only half of which is shown, and the in-plane wave vectors $k_\parallel$ along ΓK are indicated by solid arrows. The dashed spanning vector is not a calliper but conserves $k_\parallel$.

On comparing the experimental value of the long period with the values in Table I, it appears that the long period can stem from the calliper of both the spin-up and spin-down FS. From the theoretical point of view the preferred spin direction seems to be spin down. This is because in the free electron approximation which is approximately valid for the electrons responsible for the long period, the height of the potential steps, and therefore the reflection, at the Cu/Co, Co/Ni, and Ni/Co interfaces is larger (yet still small, yielding transmissions $\simeq 1$) for spin-down electrons. However, a complication arises as a result of destructive and constructive interferences in the 4 Å Co layer which can promote a certain spin direction. For the short period a calliper of the Ni FS only exists for spin-up electrons and the spin direction is clear, indeed the value for spin-up electrons agrees very well.

Let us now turn attention to the question of the conservation of $k_\parallel$. For reasons of symmetry at $k_\parallel = 0$ callipers of the FS always exist if the direction perpendicular to the surface is along a high symmetry axis, as for fcc Ni(100) and fcc Cu(100). Therefore, the condition of the conservation of $k_\parallel$ is automatically satisfied for these callipers, which are responsible for the long period in this experiment. Similarly, the condition of conservation of $k_\parallel$ is satisfied for all the experiments in the literature mentioned above, except for one which we will discuss below. In the case of the short period this cannot be decided so easily. Although the reported values of $k_\parallel$ of the callipers responsible for the short period of the Ni FS (0.50–$0.57\Gamma K$) overlap the $k_\parallel$ of Cu ($0.52\Gamma K$) it is unlikely that they are exactly the same. This implies that electron states that are callipers in Ni connect to states that are not callipers in Cu and vice versa; see Fig. 3. It appears that we have to release at least one of the conditions of (i) conservation of $k_\parallel$ and (ii) extremality of the spanning vector.

In order to determine which condition is violated, the origin of these conditions must be known. The extremality of the spanning vector is required because, in the summation over all spin density waves with perpendicular wave vectors ranging from 0 up to the Fermi wave vector, only the wave of the calliper is not cancelled. On the other hand the conservation of $k_\parallel$ is a result of the perfect in-plane translation symmetry. In practice however, to some extent interface roughness and misfit dislocations occur, resulting in a small spread in $k_\parallel$ of $\sim 1\%$.[19] Furthermore, for finite thicknesses a variation of the calliper is allowed and at elevated temperatures also a variation of $k_\parallel$. For example, from the calculated ASW band structures the spread in the calliper at room temperature was estimated 1%. A similar variation for $k_\parallel$ can be expected. These variations would almost reconcile the $k_\parallel$ of the short period of Cu with that of Ni; see Table I. Finally, as the experiment only provides us with periods, the consequences of the conservation or nonconservation of $k_\parallel$ must be translated into a period. In Table I we tabulated the calculated noncalliper and related period of the Ni FS at the $k_\parallel$ of the calliper of the Cu FS (dashed arrow in Fig. 3). The period hardly deviates from the period corresponding to the calliper of the Ni FS and we cannot establish in this way whether the extremality or the $k_\parallel$ conservation condition is violated.

Up to now the role of the thin Co layer has not been considered. The presence of callipers in Co at the same $k_\parallel$ as in Cu or Ni is not necessary for the observed oscillations. One only needs to consider the FSs of the interlayer and the layer that is varied in thickness, in this case the embedded Ni layer.[11] As long as further layers separating the aforementioned layers transmit the Bloch waves at the respective $k_\parallel$, they are unimportant in the process of selecting the callipers responsible for the observed periods. In the case of Co a band gap for spin-down electrons of the short period exists. Therefore, even if a short period calliper in the spin-down FS of Ni would exist, its contribution would be strongly reduced since tunneling is the only way of transmission.

In this respect it is worth addressing one recent cap layer experiment, where an oscillatory behavior of the interlayer coupling as a function of a cap layer was observed in Fe/Au(100).[7] In this experiment a short period oscillation resulting from a calliper at the ‘‘neck’’ of the Au FS with $k_\parallel \neq 0$ appeared. As in our experiment, the role of the Fe layer is merely to transmit the spin density waves and its FS does not play a role in the selection of callipers of the Au FS. Therefore, the conditions of the conservation of $k_\parallel$ and of the extremality of the spanning vectors at the same $k_\parallel$ are, of course, satisfied if the interlayer and cap layer are made of the same material. The cap layer type experiments can be modified to further investigate the question of conservation of $k_\parallel$ by choosing different cap layer materials than the interlayer material, e.g., Au/Fe/Ag.

In conclusion, a biperiodic behavior of the coupling strength with the thickness of an embedded Ni layer has been observed. The observed long and short periods can be related to callipers of the Ni FS. Although we cannot be conclusive due to uncertainties in both experimental and theoretical values, it is not expected that the callipers of the Cu and Ni FS share the same $k_\parallel$ in the case of the short period. Therefore the observation of a short period oscillation of the coupling across Cu as a function of the Ni thickness implies that in practice the requirement of the conservation of $k_\parallel$ or of the extremality of the spanning vector of the FS are not that strict.

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