First principles studies of modulated Co/Cu superlattices with strongly and weakly exchange biased Co-monolayers

S. Krompiewski†, F. Süss* and U. Krey*†

† Institute of Molecular Physics, P.A.N., Smoluchowskiego 17, PL-60-179 Poznań, Poland
* Institut für Physik II, Universität Regensburg, D-93040 Regensburg, Germany

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Abstract.
First-principles calculations have been performed in order to determine effective exchange integrals between strongly and weakly exchange-coupled Co monolayers in certain modulated periodic CoCu$_2$/CoCu$_n$-type superlattices with three non-equivalent Co planes, which have not yet been studied hitherto. For $3 \leq n \leq 6$ we find that the two non-equivalent exchange integrals have opposite signs, i.e. the strong coupling is antiferromagnetic and the weak coupling ferromagnetic, and differ for $n \neq 4$ from each other by one order of magnitude. It is shown that the results depend on the system as a whole and could not be obtained from separate parts. Finally we suggest that ”spin valve” systems of such kind should be considered when trying to obtain good magneto-resistance together with low switching-fields.

Magnetic multilayers based on magnetic transition metals with nonmagnetic spacers have been intensively studied for almost five years now, after it was realized that they reveal unusual oscillatory behaviour of the exchange coupling and magnetoresistance [1]. The oscillatory phenomena have a universal character, do not depend drastically on the kind of metals involved [2] and occur both with the spacer thickness as well as magnetic-slab thickness variations [3, 4, 5], and even depend sensibly on the thickness of an additional non-magnetic cap-layer on top of a Co/Cu/Co trilayers system, e.g. [6]. The explanation of these phenomena e.g. by the essentially equivalent quantum confinement, [7] and Fabry-Perrot like electron reflection theories [8], are also by now clear, and not at the center of our paper. However, recently a great deal of attention has been attracted by exchange-biased ”spin-valve” systems of the type $AF/F_1/S/F_2$ [9], with one ferromagnetic slab ($F_1$) strongly coupled to

† corresponding author, FAX xx49 941 943 4544, e-mail: krey@rphs1.physik.uni-regensburg.de
an antiferromagnet (AF) (e.g. MnFe, CoO or NiO) and the other slab (F₂) – almost free – only weakly coupled to the first one via the spacer (S). Systems of this type are not only interesting for fundamental aspects but may also be applied in future magnetic recording devices; in particular, in this way one hopes to obtain systems, where the spin direction of the weakly coupled layer is easily flipped (which explains the name "spin valves"), while at the same time the resistance of the system is sensibly changed by the flipping, which is the magneto-resistive effect mentioned below.

The purpose of the present letter is, to study by reliable ab-initio-calcuations the question, to which extent it would be possible to replace the exchange-biasing unit AF in the above-mentioned conventional "spin valve system" by a trilayer ferromagnet₁/Spacer/ferromagnet₂ involving e.g. only ferromagnetic Co planes, and Cu as spacer, provided the thickness of the spacer is chosen such as to ensure strong antiferromagnetic coupling of the two ferromagnetic layers [10].

In an attempt to get more insight into the nature of exchange coupling and possible magnetic phases in such novel systems (see below), we have studied systematically by the spin-polarized ab initio LMTO-ASA method (linearized muffin-tin orbitals, atomic sphere approximation, in the scalar-relativistic version, see [5]) the series of modulated periodic multilayers with supercell \((Co^{(2)}Cu_2Co^{(1)}Cu_2Co^{(2)}Cu_nCo^{(3)}Cu_n)\) of the (001) face-centred tetragonal structure (i.e. Co is grown epitaxially on Cu). These systems are quite specific and have never been studied before, according to our knowledge: They contain three non-equivalent Co-planes, and as we will see below, their properties cannot simply be obtained from the behaviour of conventional \((1-Co/n-Cu)\) multilayers, which we also have studied for comparison.

In our modulated systems, the monolayers \(Co^{(1)}\) couple strongly antiferromagnetically with \(Co^{(2)}\), while the \(Co^{(3)}\) monolayers turns out to be only weakly coupled (for \(n > 2\)). One of the relevant questions is the sign of this coupling (see below). The reader should note further that for computational convenience our systems are infinite multilayers, i.e. the above-mentioned supercell is periodically continued.

We have built our novel structural models on a similar basis as the simpler conventional models in our earlier papers [3, 4], in particular the in-plane atomic spacings are assumed to be equal to those of the \(fcc-(001)\) Cu with the lattice constant \(a = 3.615\) Å. Our main task has been to determine both the strong exchange coupling \(J\) between \(Co^{(1)}\) and \(Co^{(2)}\) as well as the weak coupling \(j\) between \(Co^{(2)}\) and \(Co^{(3)}\) from accurate total energy band calculations for all the relevant spin configurations, namely:

\[
(i) \quad (Co^{(2)} \downarrow Cu_2 Co^{(1)} \uparrow Cu_2 Co^{(2)} \downarrow Cu_n Co^{(3)} \downarrow Cu_n)_{∞} \quad ([↓↑↓, ↓]) \\
(ii) \quad (Co^{(2)} \downarrow Cu_2 Co^{(1)} \uparrow Cu_2 Co^{(2)} \downarrow Cu_n Co^{(3)} \uparrow Cu_n)_{∞} \quad ([↓↑↑, ↑]) \\
(iii) \quad (Co^{(2)} \uparrow Cu_2 Co^{(1)} \uparrow Cu_2 Co^{(2)} \uparrow Cu_n Co^{(3)} \uparrow Cu_n)_{∞} \quad ([↑↑↑, ↑])
\]

Obviously, since in the present studies no anisotropy is included, all the systems are spin-rotationally invariant, and there is no distinction whatsoever between the above mentioned configurations and the ones with all the spins rotated simultaneously by an arbitrary angle. After having computed the total energies of the above configurations \((E_1, E_2 \text{ and } E_3)\) the corresponding above-mentioned Cu-mediated weak resp. strong exchange coupling integrals
have been directly found from

\[ j = \frac{1}{4}(E_2 - E_1)/A, \quad J = -\frac{1}{4}(E_3 - E_1)/A, \]  

(1)

where \( A \) is the cross-section area of the unit supercell and \( E_i \) are the energies per supercell in the above-mentioned states. Furthermore, one factor of \( \frac{1}{2} \) in eqn. (1) comes from the fact that there are two thick spacers (related to the weak exchange coupling \( j \)) and two thin spacers (related to the large one, \( J \)), whereas the other factor of \( \frac{1}{2} \) results from the spin flip process according to the well known Heisenberg interaction energy per "bond" \(< ij >\):

\[ E_{<ij>} = -J_{ij} \frac{\vec{S}_i \cdot \vec{S}_j}{||\vec{S}_i|| ||\vec{S}_j||}. \]  

(2)

For comparison, we have also calculated the single exchange-integral \( j' \) for the conventional (1-Co/n-Cu)\(_\infty\) superlattices with the same program, also in scalar-relativistic version, obtained from the total energy difference \((E_{\uparrow\downarrow} - E_{\uparrow\uparrow})/(4A)\) analogously to eqn. (1).

The results of our study are presented in Fig. 1 and Fig. 2. For the novel modulated structure it can be seen that the computed couplings \( j \) and \( |J| \) oscillate with the Cu spacer thickness in a similar way. The oscillation of \( J \) shows that the system as a whole, rather than the short spacing between the corresponding Co layers alone, determines the coupling. This is in agreement with recent experiments of deVries et al., [6], however it is not our main point: More important is that the oscillations of the strong coupling \( J \) have a large negative bias (i.e. they favour antiparallel ordering of the three Co layers of type Co\(^{(1)}\) and Co\(^{(2)}\); the absolute value \(|J|\) is plotted!) and typically have a much higher amplitude than the oscillations of the weak coupling \( j \). Furthermore it is remarkable that the weak coupling \( j \) remains positive (i.e. ferromagnetic) in the range considered, i.e. for \( 3 \leq n \leq 6 \). This behaviour is in contrast to the behaviour of the exchange \( j' \) in Fig.2, which strongly oscillates from positive to negative values for the range of \( n \)-values considered and has negative values - corresponding to the first antiferromagnetic maximum - in a range where \( j \) is still positive. Of course, we cannot exclude that for \( n \geq 7 \) also \( j \) could become negative, which might be welcome for applications, and could show oscillations with \( n \) with similar periods as those of \( j' \). We can also not exclude that without our periodic boundary conditions, the weak exchange \( j \) may be negative for \( n \leq 6 \). All this would not change the conclusions from our study (see below).

In any case, for \( n = 5 \) and 6, see Fig.2, by comparison with Fig.1 we find that additionally the typical magnitude of \( j \) (Fig.1) is significantly smaller than that of \( j' \) (Fig.2). I.e. in contrast to \( j' \), the coupling \( j \) has the correct order of magnitude (except of the case \( n = 4 \), see below) when compared with experimental results on similar systems (e.g. [11]). We stress this result, since hitherto ab-initio-calculations of the present type usually gave by an order of magnitude larger amplitudes than the experiments (see [11, 12]). As already mentioned, the point with \( n = 4 \) is an exception, but even in that case \( j \) is still by a factor 0.59 smaller then \(|J|\). The reason for the peculiar behaviour at \( n = 4 \) may be some kind of reflection- or confinement-resonance, by which the antiferromagnetic state (ii) from above is more disfavoured than for \( n = 3, 5 \), and 6, compared with the ferrimagnetic arrangement (i). In any case one should
note that $j(n)$ is ferromagnetic for $n \leq 6$ at least, whereas the corresponding quantity $j'(n)$ in Fig.2 would be antiferromagnetic (i.e. $< 0$) for $n \leq 4$, and for $n = 5$ would have positive values much larger than $|j(n)|$ for any $n$. Further, in Fig. 2, $j'(n)$ oscillates clearly with a period of $\Delta n \approx 5$, whereas in Fig.1 the strong coupling $J$ seems to oscillate with a period of only $\Delta n \approx 2$. In contrast, for the weak coupling $j$ in Fig. 1, a period cannot be deduced from our data.

Thus we have found that the exchange-biasing slab $Cu_2Co^{(1)}Cu_2$ influences the coupling $j$ (between $Co^{(2)}$ and $Co^{(3)}$ via $Cu_n$) in an essential way and reduces it substantially. Concerning the accuracy of our calculations it should be stated that even in the worst case the numerical convergence criteria of our self-consistency loops are still one order of magnitude better than the small energy differences of large numbers involved in the evaluation of $j$ and $j'$, so that these results are reliable.

The $j$- and $|J|$-curves in Fig. 1 separate various magnetic phases. As $j$ never crosses zero for $3 \leq n \leq 6$ it means that at least in this region and for vanishing external magnetic field, the ground state of the multilayers under consideration is the ferrimagnetic state (i) $[\downarrow\uparrow\downarrow, \downarrow]$ (up to spin-rotational equivalence), whereas for $n \geq 7$ we cannot exclude that the antiferromagnetic state (ii) has the lowest energy. In any case, the ferromagnetic state (iii) should never be the ground state of our system.

Although we did not calculate resistances in the different configurations, and although again the results should depend on the system as a whole, we expect the following properties, which might be interesting for applications: Concerning the states (i) and (ii), as usual, in the antiferromagnetic state (ii) the resistance of the multilayers should be sensibly larger, so that systems of the present kind could be interesting for applications as magneto-resistive sensors or recording heads, if one can fix the orientation of the strongly coupled "biasing Co layers" $Co^{(1)}$ and $Co^{(2)}$ (e.g. by magnetoelastic interaction with a substrate), which would still allow for an easy switching of the weakly coupled $Co^{(3)}$ layers. This magneto-resistive effect between the ferromagnetic and the antiferromagnetic configuration should be stronger for the CPP-geometry (current-perpendicular-to-plane) than for the CIP (current in plane) geometry $[2]$, but significant enough in both cases.

As already mentioned, the physics of quantum confinement, $[3]$, see also $[13]$, or Fabry-Perrot-like multiple electron reflection, $[8]$, in ultrathin films and multilayers of the present kind, is also at the origin of the exciting coupling effects, we have calculated with our extensive calculations. We believe that the present ab initio results may yield motivation to perform additional model calculations for modulated systems with the confinement approach. In this way, one would hopefully get insight also in the reasons of the peculiar "resonance" at $n = 4$, see above, and would be able to deduce an oscillation period from examination of the large-$n$ limit. Unfortunately, our own work, which involves the accurate first-principles calculation of 24 different energies for supercells with up to 20 atoms, can be hardly extended to larger $n$.

In conclusion, our ab-initio calculation with a spin-polarized LMTO-ASA method for the possible magnetic configurations of modulated $CoCu_2/CoCu_n$ superlattices of a novel type has shown the simultaneous presence of strongly and weakly exchange-biased Co monolayers for $3 \leq n \leq 6$. It has been found that the strong coupling across two Cu monolayers is
antiferromagnetic and much larger (namely by one order of magnitude for \( n \neq 4 \), and still by a factor of \( \sim 1.7 \) in the case of \( n = 4 \)) than the coupling across the thicker spacer \( Cu_n \) with \( 3 \leq n \leq 6 \), which is ferromagnetic for these \( n \) values. Furthermore, by explicit comparison with conventional \( (1-Co/n-Cu)_{\infty} \) monolayers we have shown that in our modulated system the system as a whole, and not its separate parts, determines the properties, and that the novel modulated systems behave differently. The systems considered might be interesting when trying to obtain exchange-biased spin-valve systems without antiferromagnets, where the spin configuration can be easily switched and at the same time a sensible magneto-resistive effect can be obtained.

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References

[1] S.S.P. Parkin, N. More and K.P. Roche, Phys. Rev. Lett. \textbf{64} (1990) 2304.
[2] S.S.P. Parkin, Phys. Rev. Lett. \textbf{67} (1991) 3598.
[3] P. Bruno, Phys. Rev. B \textbf{52} (1995) 411.
[4] P.J.H. Bloemen, M.T.H. van de Vorst, W.J.M. de Jonge, M.T. Johnson and R. Coehoorn, Mod. Phys. Lett. B \textbf{9} (1995) 1.
[5] S. Krompiewski, F. Süss and U. Krey, Europhys. Lett., \textbf{26} (1994) 303.
[6] J.J. de Vries, A.A.P. Schudelaro, R. Jungblut, P.J.H. Bloemen, A. Reinders, J. Kohlhepp, R. Coehoorn, and W.J.M. de Jonge, Phys. Rev. Lett.\textbf{75}, 4306 (1995).
[7] D.M. Edwards, J. Mathon, R.B. Muniz, M.S. Phan, Phys. Rev. Lett. \textbf{67}, (1991) 493
[8] P. Bruno, J. Magn. Magn. Mater. \textbf{121} (1993) 248; Europhys. Lett. \textbf{23} (1993) 615
[9] B. Dieny, V.S. Speriosu, S. Metin, S.S.P. Parkin, B.A. Gurney, P. Baumgart and D.R. Wilhoit, J. Appl. Phys. \textbf{69} (1991) 4774.
[10] P.J.H. Bloemen, R. van Dalen and W.J.M. de Jonge, J. Appl. Phys. \textbf{73} (1993) 5972.
[11] S. Krompiewski, F. Süss and U. Krey, J. Mag. Mag. Mat. \textbf{149} (1995) I.251.
[12] K.M. Schep, P.J. Kelly, G.E.W. Bauer, Phys. Rev. Lett. \textbf{74} (1995) 586
[13] S. Krompiewski, J. Magn. Magn. Mater. \textbf{140-144} (1995) 515
Figure Captions

Fig.1: Exchange interactions $J$ and $j$ for the modulated $(CoCu_2CoCu_2CoCu_nCoCu_n)_\infty$ superlattices with strongly and weakly exchange-biased Co monolayers: The \textit{strong} antiferromagnetic exchange coupling ($J < 0$; dotted line) acts between two Co monolayers separated by just two $Cu$ monolayers ($Cu_2$), whereas the \textit{weak} ferromagnetic coupling ($j > 0$) dashed line) occurs across $Cu_n$ with $3 \leq n \leq 6$. According to eqn. (1), $j$ and $|J|$ are $\propto (E_2 - E_1)$ and $(E_3 - E_1)$, respectively, where $E_3$, $E_2$ and $E_1$ refer to the spin configurations (iii) (= ferromagnetic state, highest in energy), (ii) (= antiferromagnetic state, second highest in energy for $3 \leq n \leq 6$) and (i) (=ferrimagnetic state) sketched in the text.

Fig.2: Exchange interaction $j'$ for the conventional $(1-Co/n-Cu)_\infty$ multilayers. Note the different behaviour of $j'$ when compared with $j$ in Fig.1. The line joining the calculated points is only a guide to the eye.
$j' \text{ [erg/cm}^2\text{]}$

$\Delta (Co_1Cu_n)_\infty$

$n$

0

2 3 4 5 6

-15 -10 -5 0 5 10 15