THE INDIRECT EXCHANGE COUPLING BETWEEN FE- AND TB-LAYERS, SEPARATED BY NON MAGNETIC METAL SPACER LAYERS

Rüdiger Scherschlicht and Horst Hoffmann

Institut für Experimentelle und Angewandte Physik, Universität Regensburg,
93040 Regensburg, Germany

(Received; May 19, 1998 Accepted; August 26, 1998)

Fe/Tb multilayer films show a ferrimagnetic behaviour. The magnetic properties belonging to these systems, like the magnetic anisotropy or the saturation magnetization, can be varied by an thickness modulation of the single layers. The ferrimagnetism has the origin in the antiparallel coupling of two magnetic sublattices. The first sublattice is generated by the ferromagnetic Fe-layer and the second one originates from polarized paramagnetic Tb-layers. This ferromagnetic polarization of the Tb-layer is caused by an indirect long range exchange coupling between the Fe- and Tb-layers. For a detailed study of the named interaction the Fe- and the Tb layers were separated by a non magnetic metal interlayer (like Pt, Au, Cu or Ta) was observed. The sign of the coupling depends on the interlayer metal. In the case of Pt, Ta and Cu interlayers an antiparallel coupling of the magnetic moments of the Fe and Tb layers was observed. In the case of Au interlayers the coupling provides parallel orientation of the magnetic moments of the Fe layers and the induced moment of the Tb layers. In both cases of coupling the net magnetic moment of the multilayer system oscillates with increasing thickness of the non magnetic interlayer. The length of the oscillation period depends on the material of the non magnetic metal interlayers.

Key words: magnetism, polycrystalline multilayer films, non magnetic metal spacer layer, interlayer coupling, RKKY-interaction

1. Introduction

More and more for the last years the exchange interaction between two ferromagnetic layers via a non magnetic metal interlayer is of increasing interest. A long range coupling for example is responsible for the giant magnetoresistance in Co/Cu multilayer systems or such an interaction is also accountable for antiferro- or ferromagnetic behaviour in Fe/Cr films. At these magnetic systems a oscillatory coupling in sign and amount was observed^(1, 2, 3, 4). The oscillation amplitude of the coupling decreases with increasing thickness of the non magnetic spacer layer.

Our investigations were concentrated on the coupling of Fe/Tb multilayers. The Tb layers were paramagnetic at room temperature. Fe/Tb multilayers are surprising by

showed properties of ferrimagnetic material. All investigated multilayer films were sputter deposited. From a temperature dependence of the net magnetization and magnetization measurements on Fe/Tb-multilayer films with different Fe- and Tb-layer thicknesses we concluded a hypothesis of a long range coupling in Fe/Tb multilayer films. To confirm this model a spacer layer (Pt, Ta, Cu and Au) was inserted between the Fe and Tb layers. The thickness of the different interlayers was varied in nominal 0,1nm steps in an interval between 0,5nm and 4,5nm, depending on the interlayer element (table 1). In all multilayer systems the thicknesses of the ferromagnetic Fe layers, t_{Fe} , and of the Tb layers, t_{Tb} , were kept constant at $t_{Fe} = 1,5$ nm and $t_{Tb} = 2,2$ nm, respectively.

Each of the layers of these multilayer systems is in the thickness range of only a few monolayers. Can we expect separated layers with clear interfaces or is the whole system an alloy? Very careful investigations in the structure of the multilayer systems have been carried out. For this purpose one has to evaluate the crystalline state and the growth properties of the single layers as well as of the total multilayer films. We used element sensitive AES- and XPS depth profiles, investigations by transmission electron microscopy (TEM) of cross sections, electron scattering, and short and wide angle x-ray-diffraction (XRD).

The result of all of the structural investigations shows a very clear separation of the single multilayer periods represented by one Fe and one Tb layer separated by interlayers X like Fe/X/Tb/X (Fig. 1). All multilayer films show a very high coherence of the different layer periods and a distinct two dimensional plain growth (Fig. 2).

2. Experimental, Structure

The multilayers were sputter deposited onto glass substrates. The base pressure was $6,5 \cdot 10^{-8}$ hPa, the Ar-pressure during sputtering was $8,0 \cdot 10^{-3}$ hPa. We took special care that the base pressure before and after the deposition process was the same to cup the adsorption pollution during the deposition process on the same level. As oxidation protection all multilayer films were covered by a 7,0 nm thick layer of the interlayer material. The growth rate was kept at 0,04 nm/sec.

The number of periods was always 12. The multilayer system is shown in Fig. 1.

Fig. 1: Design of (Fe/X/Tb) multilayer films with X: Pt, Ta, Cu and Au interlayer and a cover layer. Each multilayer film contains 12 periods.

The thickness interval of all the different interlayer materials, varied in nominal 0,1nm-steps, is given in Table 1.

interlayer material X	thickness interval [nm]
Pt	0,5 – 2,5
Ta	1,0 – 4,0
Cu	2,0 – 4,0
Au	2,0 – 4,5

Table 1: Thickness array of all interlayers x in Fe/x/Tb/x multilayer films

In the following we will use the nomenclature (Fe/X:yy/Tb)₁₂ for the particular Fe/x/Tb/x multilayer films. X indicates the interlayer element and yy is the according thickness given in nm. The number 12 represents the number of periods.

The layer thicknesses were determined from the sputter time, calibrated by x-ray fluorescence and stylus measurements. The error in the layer thickness is less than 5%.

Fig. 2 shows, as an example, a typical TEM cross section image of a Fe/Tb multilayer film with nonmagnetic Au interlayers. Qualitatively, a very high coherence of the single layer periods can be seen. Also a distinct separation of all layers is obviously apparent.

Fig. 2: TEM cross section image of a (Fe/Au:4,0/Tb)₁₂ multilayer film

The XPS elemental depth profile of a Fe/Ta/Tb/Ta multilayer film, as given in fig. 3 as an example, shows, within the resolution limit of this method, very clearly the separation of the layers with distinctly separated peaks due to the multilayer structure. Each minimum of the Ta curve is correlated with a maximum of the Fe or the Tb curve. This relationship is typical for a clear separation of all single layers. With a decreased interlayer thickness this distinct separation of the single layer diminishes because of partly intermixing at the interfaces. A quantitative discussion is limited by the depth resolution of the XPS measurements. The intermixing tendencies of these multilayer systems is denoted in Table 2.

Fig. 3: XPS depth profile of a (Fe/Ta:4,0/Tb)₁₂ multilayer film

multilayer system	diffusion barrier	intermixing trend
(Fe/Pt/Tb)	Fe layer	Tb ⇒ Pt
(Fe/Cu/Tb)	Fe layer	Tb ⇒ Cu
(Fe/Au/Tb)	Fe layer	Tb ⇒ Au
(Fe/Ta/Tb)	Tb layer	Fe ⇒ Ta

Table 2: Disposition of intermixing tendency in (Fe/x/Tb) multilayer systems

The main result of the structural investigations was a clear separation of the single periods. A second result the very plain growth of the layers across a wide area. As last result we can resume, that all layers were a polycrystalline state as shown by electron and X ray diffraction.

3. Experimental, Magnetic Properties

The magnetization loops were measured at room temperature by means of a VSM in fields up to 60 kOe (exception for Cu-interlayer: $H_{\max} = 16\text{kOe}$). Torque curves were taken in fields of 16kOe. Perpendicular anisotropy was not observed. The anisotropy is only due to shape anisotropy. The characteristic magnetization loops for the four types of multilayer samples with interlayers Pt, Ta, and Au are given in figs. 4a, b, c and d, respectively. The loops were taken with fields parallel and perpendicular to the film plane. Multilayers with Pt, Ta and Cu interlayers (fig. 4a, b and c) could not

be saturated in fields up to 60 kOe. Independent of the thickness of the interlayers, the shape was always the same. The slope of the magnetization curve at large fields is much too strong to count for paramagnetism of the Tb layers. The

multilayer system behaves like a ferrimagnet, which means in a first approximation, the Fe layers and the Tb layers couple about antiparallel to each other. The magnetization loop

Fig. 4: Characteristic magnetization loops for (a) (Fe/Pt:1,7nm/Tb) -, (b) (Fe/Ta:2,6nm/Tb) -, (c) (Fe/Cu:2,4nm/Tb) - and (d) (Fe/Au:3,6nm/Tb) multilayer films. Layer thickness Fe: 1.5nm, Tb: 2.2nm. Notice: different scales for magn. moments!

Fig. 5: The net magnetization vs. the interlayer thickness of $(\text{Fe}/x/\text{Tb})_{12}$ multilayer films with (a) Pt-interlayer, (b) Ta-

of the multilayer system with Au interlayers is, independent of the interlayer thickness, of quite different shape (fig. 4d). The magnetization loop saturates. If any, only a small increase at increasing fields can be observed. This system behaves like a ferromagnet

In a first estimate it can be assumed that for Pt, Ta and Cu interlayers the observed net magnetic moment at $H=0$ is given by the difference of the moments of the Fe and Tb layers $|\vec{m}_{\text{net}}| = |\vec{m}_{\text{Fe}}| - |\vec{m}_{\text{Tb}}|$. For Au interlayers, the observed net magnetic moment is the sum of the layer moments $|\vec{m}_{\text{net}}| = |\vec{m}_{\text{Fe}}| + |\vec{m}_{\text{Tb}}|$. This assumption is caused by the hypothesis that the magnetic moment m_{Tb} of the Tb layers is different in all studied cases. This ferromagnetic behaviour of (Fe/Au/Tb) multilayer systems is surprising and can not be explained so far by the familiar rare earth-transition metal-theory^(10, 11). Hund's rule explains the coupling behaviour of these systems with the result of the well known antiparallel spin alignment of magnetic moments of Fe and Tb.

One should expect that with a increasing interlayer thickness the net magnetic moment of the multilayer systems the saturation magnetization ($M_s(\text{Fe-bcc})=1707\text{G}$, $T=300\text{K}$) because of total decoupling of the Fe- and the Tb-layers. Instead, in our investigations we found an oscillating of the net moment depending on the interlayer thickness. The net moment was determined from the backward extrapolation of the in-plane magnetization loop at zero applied field. Fig. 5 shows the dependence of the net magnetization of the four multilayer systems on the thickness of the nonmagnetic metal interlayers (fig. 5a: Pt; fig. 5b: Ta; fig. 5c: Cu; fig. 5d: Au). In all cases the net magnetization oscillates with increasing interlayer thickness. The oscillation periods are $t_{\text{osc.Pt}} = 0.5\text{nm}$, $t_{\text{osc.Ta}} = 0.8\text{nm}$, $t_{\text{osc.Cu}} = 0.6\text{nm}$ and $t_{\text{osc.Au}} = 0.7\text{nm}$. In the Ta-system (fig. 5b) more than one oscillations can be assumed.

Comparing with the saturation magnetization of the Fe (bulk material) the net magnetization of the (Fe/Pt, Ta, Cu/Tb) multilayer system is reduced. In the Ta system the decrease of the net magnetization is even stronger (fig. 5b) because of a nearly total compensation of both magnetic sublattices. But, the net magnetization of the Au system is increased. This observation agrees with the shapes of the magnetization loops, given in fig. 4, from which a ferrimagnetic and ferromagnetic coupling could be concluded, respectively.

In addition, torque curves were taken for all systems. The net magnetization was also evaluated from the torque curves⁽¹¹⁾.

The net magnetic moment was evaluated from the torque curves (TOM) and compared with the results, extrapolated from the magnetization loops (VSM). This means, two measurements completely independent of each other have been carried out. Figure 6 gives the results for multilayer systems with Pt interlayers (fig. 6a) and with Au interlayers (fig. 6b). The full curves were evaluated from the magneti-

zation loop, while the dotted curves follow from the evaluation from the torque measurements.

In both cases the oscillation of the net magnetic moment with increasing thickness of the Pt and Au interlayer shows the same periods, $t_{\text{osc.Pt}}=0,5\text{nm}$ and $t_{\text{osc.Au}}=0,7\text{nm}$, respec-

Fig. 6: Comparison of the net magnetic moment taken from VSM (straight line) and TOM (dotted line) measurements for (a) (Fe/Pt/Tb)₁₂ multilayer films and (b) (Fe/Au/Tb)₁₂ multilayer films

tively.

The maxima of the oscillations, obtained from the magnetization loops and from torque measurements coincide for Au interlayers. While for Pt – interlayers the maximum of the oscillation obtained from torque measurements coincides with the minimum of the oscillation, determined from the magnetization loops. In the first case the Fe – Tb layers are ferromagnetically coupled, while in the latter case they are ferrimagnetically coupled, as can be seen from the magnetization loops. From the measurements we conclude, that in each case a strong constant coupling is superimposed by weaker oscillating coupling. The coupling strength determines the magnetic moment of the Tb layers, depending on the thickness t of the interlayers.

$$m_{\text{Tb}}(t) = m_{\text{Tb}}^0 (1 + f(t))$$

The oscillation is given by $f(t)$ with $|f(t)| < 1$. The total magnetic moment of magnetization loop is given by

$m = m_{\text{Fe}} \pm m_{\text{Tb}}$. The sum represents ferromagnetic coupling, the difference gives ferrimagnetic coupling.

The maximum of the torque is given by $L = B(m_{\text{Fe}}^2 + m_{\text{Tb}}^2)$ with $B = \text{constant}$. From magnetization loops one obtains

$$m = m_{\text{Fe}} \pm m_{\text{Tb}}^0 (1 + f(t)) .$$

From torque curves one evaluates

$$L = B\{m_{\text{Fe}}^2 + (m_{\text{Tb}}^0)^2 (1 + f(t))^2\} .$$

The maxima are given by

$$\frac{dm}{dt} = \pm m_{Tb}^0 \frac{df(t)}{dt} = 0, \quad \frac{dL}{dt} = 2B(m_{Tb}^0)^2(1+f(t))\frac{df(t)}{dt} = 0$$

and

$$\frac{d^2m}{dt^2} = \pm m_{Tb}^0 \frac{d^2f(t)}{dt^2} < 0 \quad (1)$$

$$\frac{d^2L}{dt^2} = 2B(m_{Tb}^0)^2 \left(\frac{df(t)}{dt} \right)^2 + 2B(m_{Tb}^0)^2(1+f(t))\frac{d^2f(t)}{dt^2} < 0$$

Since $\frac{df(t)}{dt} = 0$ at the maximum, one obtains

$$\frac{d^2L}{dt^2} = 2B(m_{Tb}^0)^2(1+f(t))\frac{d^2f(t)}{dt^2} < 0 \quad (2)$$

Comparing eqs(1) and (2) one finds that for ferromagnetic

coupling both maxima occur at $\frac{d^2f(t)}{dt^2} < 0$.

For ferrimagnetic coupling the magnetization loop measurements leads to $\frac{d^2f(t)}{dt^2} > 0$ (minimum).

While the torque measurements leads simultaneously to

$$\frac{d^2f(t)}{dt^2} < 0 \quad (\text{maximum}).$$

This explains the shift of the oscillation curves in the case of the ferrimagnetic coupling and the non-shift for ferromagnetic coupling.

4. Discussion

The results show the existence of a long range indirect exchange interaction between Fe- and Tb-layers, even if layers are separated by non magnetic metal interlayers. The strength and the sign of the coupling depends on the material of the interlayers. In any case a magnetic order is induced into the Tb layers. It can be assumed that the Fe layers originate a coupling field by indirect exchange interaction in the paramagnetic Tb layers. The coupling field is directed antiparallel to the Fe moment in the case of Pt, Ta and Cu interlayers and parallel in the case of Au interlayers. With this assumption the observed oscillation of the net magnetic moment is explained by an oscillation of the exchange field. The oscillation period seems to be connected RKKY-exchange interaction^(5, 7). Based on this theory, which is applied on magnetic multilayer films, the long

range behaviour of coupling oscillations can be declared as the positive sign of the net magnetization, too. In (Fe/x/Tb) multilayer systems each magnetic element (Fe and Tb) generates a magnetic sublattice. Only the non intermixed magnetic layer with plain interfaces (see table 2: diffusion barrier) is able to be responsible for a long range oscillating exchange interaction like a RKKY-exchange interaction. The second magnetic sublattice indicates a magnetic off-set, which hold the net magnetization always on a positive level and thereby prevents that the magnetic properties change periodically from a ferro- to a antiferromagnetic behaviour.

References

- [1] R. Coehoorn: Phys. Rev. B **44**, 9331 (1991).
- [2] C. Carbonne and S. F. Alvarado: Phys. Rev. B **36**, 2433 (1987).
- [3] A. Fert and P. Bruno: Ultrathin Magnetic Structures II ed. by B. Heinrich and J. A. C. Bland, p. 82 (Springer-Verlag, Berlin 1994).
- [4] S. S. P. Parkin, K. P. Roche and T. Suzuki: J. Appl. Phys. **74**, 5886 (1993).
- [5] C. Chapert and J. P. Renard: Europhys. Lett. **15**, 553 (1991).
- [6] P. Bruno and C. Chappert: J. Appl. Phys. **67**, 1602 (1991).
- [7] P. Bruno and C. Chappert: Phys. Rev. B **46**, 261 (1992).
- [8] Z. Shi, P. Levy and J. L. Fry: Phys. Rev. B **49**, 15159 (1994).
- [9] M. S. S. Brooks, L. Nordström and B. Johansson: J. Phys. Condens. Matter **3**, 2357 (1991).
- [10] M. S. S. Brooks, T. Gasche, S. Auluck, L. Nordström, L. Severin, J. Trygg and B. Johansson: J. Appl. Phys. **70**, 5972 (1991).
- [11] G. Pastor, A. Ferreira and M. Torres: J. Magn. Magn. Mater. **53**, 354 (1986).