Investigation of the as-deposited characteristics and the thermal stability of CoFe and Ru based multilayers

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The as-deposited characteristics and the thermal stability at 300 °C of CoFe and Ru based ultrathin multilayers have been investigated and structural and magnetic characteristics have been correlated. These layer systems constitute the basis of the hard artificial antiferromagnetic subsystem used in current sensors. The coercive field of 2 nm CoFe (3.5 kA/m) is found to increase linearly by a factor of 13 upon increasing the Ru seed layer thickness from 5 to 40 nm. The weakening of the exchange coupling due to the sharpening of the crystallite boundaries is likely the origin. Magnetization-induced second-harmonic generation measurements show that the CoFe/Ru interface remains magnetically active after 36 h with a magnetic contrast of 0.17, indicating that no strong mixing at the interface is taking place. In accordance, the saturation field of the antiferromagnetic coupling in $\text{Ru}_5/\text{CoFe}_2/\text{Ru}_{0.7}/\text{CoFe}_2/\text{Ru}$ (nm) does not change during 20 h of annealing. However, slowly a remanent magnetization develops which is thought to be due to the formation of ferromagnetic shortcuts through the crystallite boundaries of the coupler. © 2002 American Institute of Physics. [DOI: 10.1063/1.1455617]

I. INTRODUCTION

Recently, devices based on the giant magnetoresistance (GMR) effect in magnetic multilayers have been introduced in the market, like thin film heads and GMR sensors.¹ A lot of effort has been put into the further development of these sensor² applications as well as into new applications like magnetoresistive memories.³ In general, these devices consist of a soft magnetic layer, with a magnetization that can be easily rotated, and a magnetic hard layer or hard subsystem, which basically should not respond to the external field. The so-called artificial antiferromagnet (AAF), consisting of two antiferromagnetically coupled Co layers, has gained much interest due to its successful application as a hard subsystem with relatively high magnetic rigidity.¹ This will also be of importance for the development of the next generation of devices based on the tunnel magnetoresistance effect. Important aspects for the success of these devices are the magnetic hardness of the AAF, the thermal stability of their structures and interfaces, and the flexibility or the tunability of their magnetic characteristics. For this and for further improvement, it is important to trace the critical physical processes that determine the stability and to find the design and preparational parameters that enable tuning of the characteristics. Much emphasis will be put on the correlation between the structural and magnetic characteristics. On the one hand, both the thermal stability and the magnetic characteristics of the AAF crucially depend on its single layers while on the other hand the interplay in the sensor stack adds extra critical parameters. In this article we present the results of our inves-

tigation into the complete AAF stack: glass/Ru/CoFe/Ru/CoFe/Ru/Cr as well as separate studies on isolated parts.

II. EXPERIMENT

The samples were deposited on glass substrates of $9 \times 9 \times 0.4 \text{ mm}^3$ by dc magnetron sputtering at an Ar pressure P_{Ar} of 10^{-2} mbar. For annealing, the samples, prepared in the same run, were put on a preheated base plate in ovens with atmospheric environment at a constant temperature of 300 °C. Successively, at predefined time steps the samples were taken out one by one and subsequently measured at room temperature with various techniques like vibrating sample magnetometry (VSM), magnetoresistance measurements with the standard four point method, x-ray diffraction (XRD), small angle x-ray scattering (SAXS), atomic force microscopy (AFM), transmission electron microscopy (TEM) and magnetization-induced second-harmonic generation (MSHG) measurements.

MSHG is a nonlinear magneto-optical technique that has been shown to be a powerful and simple tool to study magnetic multilayers, combining an extreme surface/interface sensitivity^{4,5} with very large magneto-optical effects. For the MSHG measurements, a pulsed laser beam from a Ti: sapphire laser (82 MHz $\times$ 100 fs pulses at 840 nm and average power of 700 mW) was focused onto the sample with a spot diameter of 100 μm . Measurements were done in the transversal configuration (for details see Ref. 4). The studied AAF-stack geometry is as follows: glass/ $\text{Ru}_5/\text{CoFe}_2/\text{Ru}_{0.7}/\text{CoFe}_3/\text{Ru}_5/\text{Cr}_5$. Here the subscripts indicate the layer thicknesses in nm. The following test structures with one magnetic layer have been used: glass/ $\text{Ru}_x/\text{CoFe}_y/\text{Ru}/\text{Cr}_5$ with $x=5, 7.5, 10, 15, 30, 45$ nm and $y=2, 4, 40$ nm, indicated by Ru_xCoFe_y . For all samples we

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FIG. 1. H_c of the Ru_xCoFe_2 samples determined with VSM measurements plotted as a function of the Ru buffer layer thickness. Three normalized hysteresis curves of samples with buffer thicknesses of 5, 15 and 45 nm are shown in the inset.

used a 5 nm Cr or 30 nm Ru cap layer to protect against oxidation. Single Ru layer samples with varying thicknesses have been made as well: glass/ Ru_x indicated by Ru_x .

III. RESULTS AND DISCUSSION

A. Tunability of the magnetic hardness

Figure 1 shows that H_c increases linearly with Ru buffer layer thickness of the Ru_xCoFe_2 samples, as determined with VSM. This enables us to tune the rigidity of the 2 nm CoFe layer in the range 3–45 kA/m. This allows the control of the magnetic hardness of the AAF subsystem since its rigidity directly depends on the stiffness of the single layers, which is experimentally confirmed. From the shape of the hysteresis curves in Fig. 1, it is obvious that the coercivity is dominated by domain-wall motion, i.e., by wall friction. In order to find the dominant cause of this friction increase, several techniques have been applied to check the following layer characteristics: interface roughness, Ru texture, Ru crystallite sizes and the degree of granularity, this all as a function of Ru thickness. AFM measurements on Ru_x samples and SAXS measurements on the $\text{Ru}_x/\text{CoFe}_2$ samples indicate a small increase in roughness with seed layer thickness, but were obscured by the error bars. The rms value obtained with SAXS for the Ru/CoFe interface was $6.5 \pm 1.5 \text{ \AA}$. XRD measurements on the Ru_x samples showed that the Ru has a polycrystalline structure where over 80% of the diffracted intensity indicated a (002) hcp texture and 15% a (101) texture. With increasing Ru thickness these fractions did not change and also the absolute intensities divided by the thickness did not change. The full width at half maximum (FWHM) of the largest peak (002) showed a decrease (sharpening) with increasing thickness, in good agreement with the Scherrer equation⁶ that relates the thickness of the crystallites to the FWHM of the Bragg peak. This implies that the Ru layer has columnar growth, which was confirmed by TEM measurements of the Ru_{45} sample (Fig. 2). Also something of a slight cone-like growth can be observed, however the increment in the crystallites diameter with thickness is too low to account for the H_c dependence.⁷ The TEM image also shows that the borders between the column-like crystal-

FIG. 2. TEM measurements of the $\text{Ru}_{45}/\text{CoFe}_2/\text{Ru}_3/\text{Cr}_5$ nm sample showing the columnar growth and the sharpening of the crystallite boundaries with increasing thickness.

lites are becoming sharper and more pronounced as the Ru layer is becoming thicker. Furthermore, coherent growth of the CoFe on Ru can be seen in Fig. 2 at different locations, and TEM measurements on $\text{Ru}_{15}/\text{CoFe}_{40}/\text{Ru}/\text{Cr}$ also showed CoFe column-like growth correlated with the Ru columns.

It is clear that changes in the Ru crystallite diameter, roughness or texture are not significant. The Ru crystallite boundaries, on the other hand, are clearly far less pronounced for small Ru thicknesses than for larger thicknesses. Combined with the observation that the CoFe crystallite growth is correlated with the Ru crystallites, it is evident that the CoFe growth will be affected by the seedlayer thickness. Considering that crystallite boundaries are preferred locations where domain walls tend to experience strong pinning forces, it is thought that the weakening of the exchange coupling at the CoFe crystallites with increasing Ru seedlayer thickness is the likely cause.

IV. THERMAL STABILITY

XRD data on $\text{Ru}_{15}/\text{CoFe}_{40}/\text{Ru}/\text{Cr}$ show the (200) and the (101) peaks of Ru and the (110) bcc peak of CoFe. Other CoFe peaks gave negligible intensities. The (110) texture of CoFe has a lattice mismatch of 8% with Ru. As the mismatches of other orientations were all much larger, the growth of this texture is not surprising. From the as-deposited XRD curves it was found that the (110) peak was shifted by 0.4% compared to its literature value, indicating that the CoFe is slightly stretched in plane.

Upon annealing, the (110) CoFe peak shifted to its literature value, while the intensity of the peak reduced, indicating that texture transformation occurred. The (200) Ru peak was found to increase in intensity while maintaining its position. The (101) Ru peak did not change at all. As the increase of the (002) peak occurred without the reduction of the other peak, it is most likely related to crystallization⁸ of amorphous or nano-crystalline Ru into the (002) texture. These changes were accompanied by a reduction of the electrical resistivity by 40% after 8 h of annealing and the reduction of the thicknesses of Ru (45 nm) and CoFe (40 nm) by about $1 \pm 0.2\%$. This indicates that upon annealing the CoFe is relaxing to a closer packed state probably due to stress relaxation and crystallization. These processes are not surprising since the sputtering technique grows the multilayer in a nonequilibrium state.

The MSHG measurements are shown in Fig. 3. The open squares are the second-harmonic intensities when the mag-

FIG. 3. MSHG curves of the $\text{Ru}_{15}/\text{CoFe}_{40}/\text{Ru}_3/\text{Cr}_5$ as a function of annealing time at 300 °C. The open squares are the second-harmonic intensities when the magnetization is pointing upward and the closed squares when the magnetization points downwards.

netization was pointing along the positive y direction (upward) and the closed squares when the magnetization was reversed. In the beginning the intensity increased strongly with annealing, thereafter it was constant within the error bar. The intensity change upon magnetization reversal (the magnetization induced contribution shifts its phase by 180°) exhibited a similar course. The initial strong increase in intensity (over 400%) shows that the interfaces are strongly affected by the annealing process. Considering that small material movements in the bulk may alter the interface dramatically, the huge changes in the MSHG intensities are not unusual. On the other hand, the fact that the magnetic contribution remained strong indicates that the CoFe/Ru interface remained magnetically active and that no strong interface mixing had occurred.⁹

In the measurements of the $\text{Ru}_{40}/\text{CoFe}_2/\text{Ru}_3/\text{Cr}_5$ sample, a decrease of R_S and M_S (18%) was found upon annealing. The H_C of the 2 nm CoFe had increased and saturated within 8 h of annealing at 300 °C. This increase implies a stronger wall pinning, which may have been caused by the reduction of the exchange coupling between crystallites due to diffusion of material into their boundaries. The stability of the MSHG magnetic signal combined with the reductions of the M_S supports the motion of material into the crystallite boundaries.

Figure 4 shows GMR curves of the AAF

FIG. 4. Four GMR curves of the AAF $\text{Ru}_5/\text{CoFe}_2/\text{Ru}_{0.7}/\text{CoFe}_2/\text{Ru}_4/\text{Cr}_5$ annealed for different times (0, 0: 15, 2:15, 20:25 h) at 300 °C. The inset shows the same curves but normalized.

$\text{Ru}_5/\text{CoFe}_2/\text{Ru}_{0.7}/\text{CoFe}_2/\text{Ru}_4/\text{Cr}_5$ annealed for different times at 300 °C. The saturation fields (H_S) here were low due to the sputtering conditions used, leading to relatively higher mixing of the interfaces and lower interlayer exchange coupling. The inset shows that the normalized curves coincided, i.e., the saturation field remained unchanged. However, the GMR slowly decreased with annealing and a net remanence magnetization revealed itself after 2:15 h (10% of M_S), increasing to 70% after 20:25 h. The occurrence of the remanence signifies the development of ferromagnetically coupled areas so that these portions no longer contribute to the GMR. Since the response of the AF fraction of the stack was not affected by the anneal, i.e., the shape was not changed significantly, the ferromagnetic coupling between layers occurs by local ferromagnetic bridges, likely located at the crystallite boundaries in the Ru. These results clearly show that the $\text{CoFe}/\text{Ru}_{0.7}/\text{CoFe}$ is not very stable and a slow but continuous degradation is occurring.

In conclusion, we have found a linear relationship between coercive field and buffer layer thickness in as-deposited samples of CoFe/Ru multilayers that is most likely due to the weakening of the exchange coupling related to the sharpening of the crystallite boundaries in the Ru. Thus, the hardness of the AAF can be tuned by adapting the Ru buffer thickness. Upon annealing, the single magnetic layer samples showed indications for structural relaxation, and limited crystallite boundary diffusion of Ru into CoFe, while the interfaces are only weakly affected. Annealing of the AAF samples at 300 °C resulted in a slow but gradual degradation due to the formation of ferromagnetic regions leading to loss in the GMR signal and to the development of a remanence. These regions originate most likely in ferromagnetic bridges along the crystallite boundaries in the Ru coupling layer. Due to the tunability of its hardness and strong interlayer exchange coupling, Ru/CoFe is a good candidate to be employed in various applications as long as the operation temperature remains well below 300 °C.

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