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Thermal stability and degradation mechanism of NiFe/Cu giant magnetoresistance multilayer systems

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$Ni_{80}Fe_{20}$/Cu multilayers show large giant magnetoresistance (GMR) at low magnetic saturation fields. The GMR signal is known to degrade irreversibly at elevated temperatures. Clarification of the relevant deterioration mechanisms refines our basic understanding of the GMR effect and may help to improve the thermal stability of devices. We therefore investigated structural, transport, and magnetic properties of sputtered $Ni_{80}Fe_{20}$/Cu multilayers in the as-deposited state and after different anneals (up to 600 °C) by x-ray techniques, transport measurements, ferromagnetic resonance (FMR), and magneto-optical Kerr effect (MOKE). Multilayers with the second maximum of the antiferromagnetic (afm) coupling showed a sharp drop of the GMR at about 250 °C. The changes of the transport properties were associated with a series of structural alterations. These ranged from grain growth and defect reduction through texture sharpening and stress evolution up to the onset of interdiffusion. Interdiffusion changed the NiFe layer composition and the interface structure and finally caused layer intermixing with a loss of the former multilayer structure. Further insight into the magnetic behavior was gained from FMR and MOKE measurements, from which we determined the in-plane magnetic anisotropies, the strength of the afm coupling (bilinear and biquadratic), and the homogeneity of the layer magnetization as a function of the annealing temperature. *© 2002 American Institute of Physics.* [DOI: 10.1063/1.1452201]

The discoveries of antiferromagnetic exchange coupling[1] and giant magnetoresistance (GMR) effects[2–4] have opened a possibility for applications in different areas, such as magnetic recording, nonvolatile memories, and magnetic sensors. One of the most important applications was the development of spin valve structures by IBM.[5] In order to optimize the performance of magnetic devices, many different material combinations have been studied to obtain optimum properties. Among them, the Ni_xFe_{1-x}/Cu ($x \approx 0.81$, in the following denoted as NiFe or Permalloy) system[6,7] has attracted significant attention due to the low anisotropy in permalloy, the small saturation magnetic field, and negligible hysteresis effects.

The performance of the magnetic devices based on the NiFe/Cu material combination must withstand different working conditions, such as elevated temperature and mechanical stress. Depending on the individual layer thickness, an irreversible degradation of the GMR occurs in NiFe/Cu multilayers at elevated temperatures.[8] However, little is known about the underlying individual deterioration mechanisms; one must perform comprehensive structural analysis in order to understand these mechanisms. For multilayers with 100 nm individual layer thicknesses and thicker, structural investigations by Auger electron spectroscopy (AES) and x-ray diffraction (XRD) indicated the onset of Ni diffusion into the Cu layers above a critical temperature.[9] The present investigation concerned the question of how the degradation of the magnetic properties is correlated with irreversible structural changes for *nanoscaled* GMR multilayers. To do this, we used a whole spectrum of methods, including x-ray diffraction and reflectometry, electron microscopy, measurements of the transport properties, magneto-optical Kerr effect (MOKE), and ferromagnetic resonance (FMR).

We employed dc magnetron sputtering to deposit $[\mathrm{NiFe}(1.7\ \mathrm{nm})+\mathrm{Cu}(2.1\ \mathrm{nm})]_{30}+\mathrm{NiFe}(1.7\ \mathrm{nm})$ structures onto thermally oxidized Si (001) wafers in an Ar atmosphere of 60 mbar. The Cu layer thickness corresponded to the second antiferromagnetic maximum in the NiFe/Cu system. We employed a Philips-XPert diffractometer with Cu K_α radiation to carry out the x-ray diffraction experiments and a standard four-point probe setup to measure the transport properties. We conducted anneals for 1 h in vacuum (pressure 10^{-6} mbar) at different temperatures (T_{an}) in the range between 75 and 600 °C. After the anneals, we carried out structural, transport and magnetic measurements at room temperature.

The GMR effect in the as-deposited structures was on the level of 10%. Annealing up to 220 °C increased the GMR to 12%, however, additional annealing (at 300 °C or higher temperatures) resulted in a sharp decrease of the GMR signal (see Fig. 1). We found that the total resistivity of the samples was nearly constant up to 220 °C and strongly increased after annealing at 300 °C. The saturation field behaved in a similar fashion, it was nearly constant (approximately 80 Oe) for annealing up to 220 °C and then increased dramatically to approximately 1500 Oe for higher annealing temperatures.

To correlate the changes in the GMR properties with possible changes of the interface properties, we used x-ray reflectometry (XRR). The XRR patterns clearly showed that

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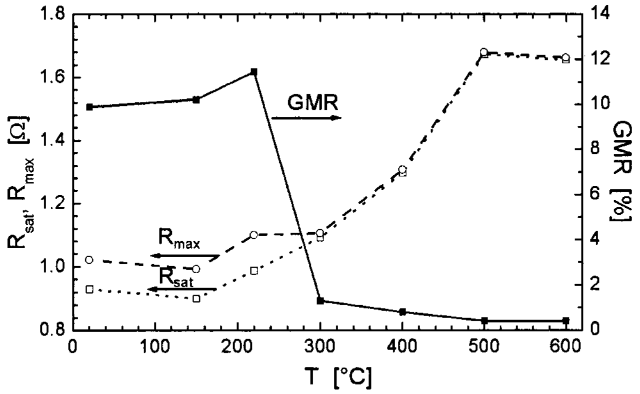

FIG. 1. Maximum resistance at zero magnetic field (R_{max}), saturation resistance (R_{sat}), and GMR vs annealing temperature.

the bilayer sequence is stable up to an annealing temperature of 300 °C.[10] For higher annealing temperatures we observed degradation of the layered structure, which completely intermixed after annealing at 600 °C. In contrast, the total thickness of the metallic structure (120 nm) was preserved even after annealing at 600 °C. From simulation calculations of the XRR curves, we calculated the mean roughness parameter (σ) of the interfaces to be 0.5 nm up to $T_{an}\sim 250\,°C$, followed by a sharp increase to values above 1 nm at $T_{an}\sim 400\,°C$.

The wide angle diffraction patterns (Fig. 2) indicated that a predominantly ⟨111⟩ texture was preserved during annealing. Even the texture sharpened during annealing, i.e., the halfwidth of the pole figure cuts decreased.[10] The grains possessed a typical vertical size of approximately 25 nm after deposition and showed a columnar structure. Annealing at $T_{an} > 220\,°C$ increased the grain size significantly, causing a growth of a certain fraction of grains through the complete layer stack, as seen also in Co/Cu multilayers.[11] Still more striking was the lateral grain growth, which lead to maximum grain sizes in the micrometer range and a mean size of 700 nm after the 400 °C anneal. This was measured using the electron back scattering diffraction technique in a scanning electron microscopy. AES measurements of NiFe (100 nm)/Cu (200 nm) stacks showed the onset of interdiffusion at 250 °C, when Ni atoms preferentially diffused into the Cu layers.[9] The x-ray and AES measurements indicated that anneals above 250 °C strongly affected the multilayer structure. The strong increase in grain size and roughness parameters, in combination with preferential interdiffusion of Ni into Cu, finally resulted in a complete destruction of the layered NiFe/Cu structure. More detailed results of the XRR and of the texture studies are presented elsewhere.[10]

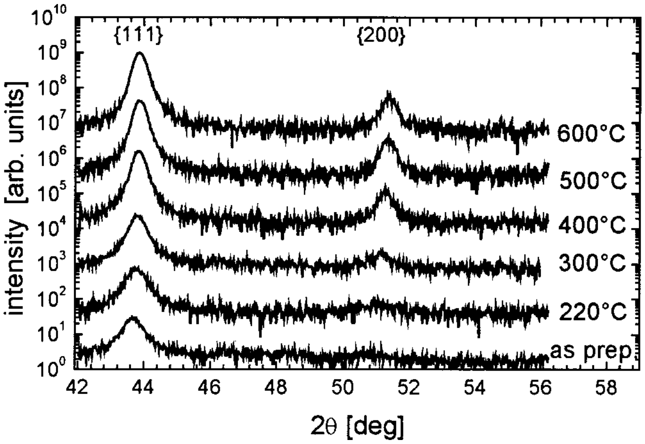

FIG. 2. Wide angle x-ray diffraction patterns showing the zeroth order {111} and {200} reflection of the NiFe/Cu multilayers.

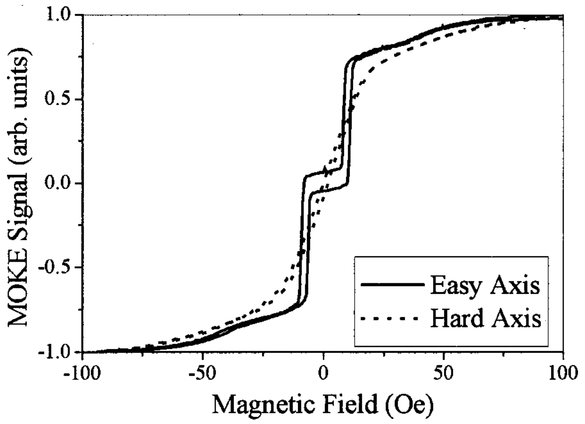

FIG. 3. The hysteresis loops measured along different axes (easy and hard).

We performed magnetic measurements at room temperature as a function of the angle within the film plane. Figure 3 shows hysteresis loops measured by MOKE for different orientations. The change in the shape of the hysteresis loops for different angles (only two shown) clearly indicated the presence of a uniaxial in-plane anisotropy. Measurements along one direction yielded "S"-shaped loops (hard axis) and for measurements 90° off this axis we observed hysteresis loops that are typical for afm coupled systems. The value of the saturation along the easy axis (approximately 75 Oe) was in agreement with the GMR measurement for the as-deposited samples. The shape of the hysteresis loop along the easy axis was typical for the presence of both biquadratic and bilinear coupling contributions. Two critical fields were seen. The first represented the initial deviation from the collinear (saturated) configuration and the second represented a field at which the neighboring layers became antiparallel. From these measurements, we determined the strength of both, the bilinear (J_1) and the biquadratic (J_2) exchange coupling[12] ($J_1 = -0.0012\,\mathrm{erg/cm^2}$ and $J_2 = 0.001\,\mathrm{erg/cm^2}$) and the strength of the small uniaxial in-plane anisotropy ($H_u = 20\,\mathrm{Oe}$). The strength of the exchange coupling was very small and nearly constant up to $T_{an}\sim 260\,°C$. At this temperature the hysteresis loops showed visible deformations with respect to the as-grown data. Instead of two well-defined critical fields we observed a few smaller jumps that indicated different switching fields in different regions of the sample.

The FMR measurements confirmed the presence of the uniaxial in-plane anisotropy and resulted in values that were similar to those determined by the fitting of the MOKE data.[13] The uniaxial in-plane anisotropy ($20\pm 5\,\mathrm{Oe}$) was nearly constant up to $T_{an}\sim 260\,°C$. Then we observed a significant increase to 60 Oe at $T_{an}\sim 330\,°C$. The $4\pi M_{eff}$ behaved in a similar fashion. Up to 260 °C, the value of $4\pi M_{\text{eff}}$ was nearly constant at 6.5 kG; this was followed by a rapid decrease to 5.6 kG at 330 °C.

The measurements of the FMR linewidth revealed an interesting behavior. The line width was nearly constant ($\Delta H = 120$ Oe) up to $T_{\text{an}} \sim 150\,°\text{C}$. After annealing at 180 °C the linewidth decreased to 75 Oe. However, after annealing at higher temperatures we observed increased values of the linewidth, which reached a maximum value of 250 Oe after annealing at 330 °C—see Fig. 4. The observed minimum of the linewidth corresponded well to the increased GMR at this annealing temperature—a finding that was discussed in more detail by Hecker *et al.*[10]

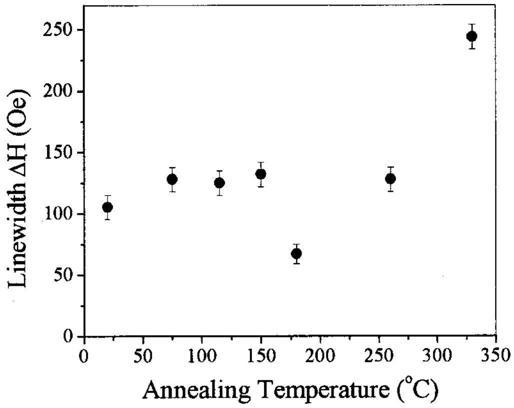

FIG. 4. FMR linewidth as a function of annealing temperature, measured at 24 GHz.

All our experimental results pointed to a temperature of approximately 250 °C, at which we found critical changes in our NiFe/Cu structures. We observed two tendencies. First, for annealing below 250 °C, there was an increase of the grain size and a reduction of defects, as inferred from the XRD experiments. This tendency corresponded to the observed decrease in FMR linewidth, which indicated increased magnetic homogeneity of our layers near an annealing temperature of 200 °C. Second, for annealing temperatures above 250 °C, we observed a significant intermixing between Ni and Cu that degraded the structural and magnetic integrity of the NiFe/Cu layers. The multilayer structure became less defined, and as a result we observed a significant degradation of the GMR effect. In conclusion, it is the alloying tendency of Ni and Cu above 250 °C that determined the decay of the GMR and the change in the magnetic properties of our NiFe/Cu multilayers.

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