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Annealing effects and degradation mechanism of NiFe/Cu GMR multilayers

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Abstract

Structural, transport and magnetic properties of sputtered $Ni_{80}Fe_{20}$/Cu multilayers showing giant magnetoresistance (GMR) were studied using X-ray reflectometry and diffraction, transport measurements, ferromagnetic resonance (FMR), and magneto-optical Kerr effect. In particular, mechanisms of the GMR degradation at elevated temperatures were investigated. Multilayers with an individual layer thickness of 2 nm show a sharp drop of the GMR after annealing at about 250°C. Whereas below this temperature grain growth and defect reduction contribute to a partial improvement of the GMR, above ~250°C interdiffusion between Ni and Cu appears to lead to layer intermixing and to the degradation of transport and magnetic properties. Moreover, the initial $\langle 1\,1\,1 \rangle$ texture sharpens, and strong tensile stresses arise in the layer stack. We correlated the structural alterations to changes in the magnetic properties such as the strength of the antiferromagnetic coupling (bilinear and biquadratic) and the magnetic anisotropy. Above 250°C an increasing magnetic inhomogeneity of the Permalloy layers can be inferred from the FMR linewidth broadening. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

Nanoscale metallic multilayers containing alternating ferromagnetic and nonmagnetic layers show dazzling magnetic and transport properties such as giant magnetoresistance (GMR), which is of great interest for applications in the field of mass storage technology, sensor development, and magnetoelectronics [1]. In particular, the Ni_xFe_{1-x}/Cu materials combination ($x \approx 0.81$, in the following denoted as NiFe or Permalloy) is attractive due to the high GMR ratios attainable at low magnetic saturation field with negligible hysteresis effects [2,3].

In contrast to alternative systems like Co/Cu which are immiscible, NiFe and Cu can show interdiffusion. For stacks with film thicknesses of 0.1 and 0.2 μm for NiFe and Cu, respectively, the

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onset of interdiffusion was observed to occur at about 200°C [4]. Thus the question arises if these effects also determine the properties of thin nanoscale layer stacks at elevated temperatures, and how they interfere with the thermal stability of the GMR multilayers. Depending on the individual layer thickness, an irreversible degradation of the GMR occurs in nanoscale NiFe/Cu multilayers at elevated temperatures [5]. The underlying mechanism is of interest for both, the fundamental understanding of the GMR effect and the thermal stability of GMR devices. During annealing different effects on the structure of thin metallic films have been observed, for example the formation of Ag bridges in NiFe/Ag multilayers [6], the break-up of Co layers in Co/Cu multilayers [7,8], the growth of pure Ag and Ni grains in Ag/Ni multilayers [9], or the texture alterations connected with abnormal grain growth in Co/Cu multilayers [10] and Ag films [11]. The goal of the present investigation is to correlate structural, transport, and magnetic properties of the Ni_xFe_{1-x}/Cu multilayers as a function of the annealing temperature, and thus to better understand the underlying mechanisms of the GMR decay. Complementary methods such as X-ray diffraction (XRD) and reflectometry, electron microscopy, measurements of the transport properties, magneto-optical Kerr effect (MOKE) and of ferromagnetic resonance (FMR) were combined to trace the changes of the multilayer structure and properties during the anneals.

2. Sample preparation and experimental techniques

The $Ni_{81}Fe_{19}$/Cu multilayer films with a total thickness of about 120 nm were deposited by dc magnetron sputtering onto thermally oxidized Si (1 0 0) wafers in an Ar atmosphere of 0.6 Pa. The nominal structure of the samples was $[NiFe(1.7\,nm) + Cu(2.1\,nm)]_{30} + NiFe(1.7\,nm)$ with the Cu thickness corresponding to the second antiferromagnetic (afm) interlayer coupling maximum. The samples were annealed for 1 h in vacuum ($p \sim 10^{-4}$ Pa) at different annealing temperatures, T_{an}, in the range between 75°C and 600°C. After annealing, structural, transport and magnetic properties were measured at room temperature.

Transport properties of the multilayers were investigated by a standard 4-point probe set-up operated at room temperature. As a measure of the GMR, the ratio $(R_{max} - R_{sat})/R_{sat}$ was determined with R_{max} and R_{sat} being the maximum resistance at zero field and the saturation resistance at high external magnetic fields, respectively. The XRD experiments were done using a Philips-XPert diffractometer with Cu-K_α radiation, Eulerian cradle and thin-film equipment. Both low- and wide-angle measurements were performed at room temperature. The magnetic properties were determined using FMR and MOKE. The FMR measurements were conducted at 10 and 24 GHz using a cylindrical cavity (TE_{011}-mode).

3. Experimental results

3.1. Magnetotransport properties

The NiFe/Cu samples showed a clear GMR effect (see Fig. 1) with a low saturation field of 80 Oe. This value of the saturation field remained unchanged within the experimental error, up to an annealing temperature of 220°C. Annealing above this temperature (300°C or higher) resulted in a significant increase of the saturation field to ~1500 Oe.

The behaviour of the GMR as a function of annealing temperature as seen in Fig. 2 shows a

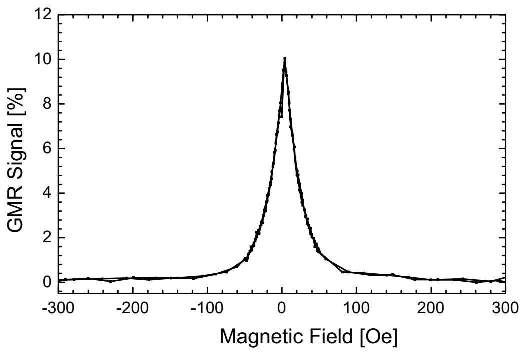

Fig. 1. Change of the resistivity as a function of the magnetic field in the NiFe/Cu structure after deposition.

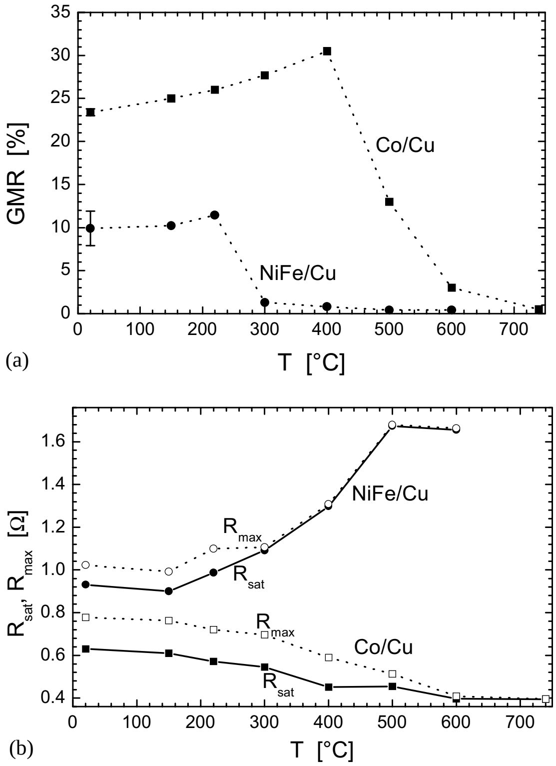

Fig. 2. (a) GMR effect as a function of annealing temperature. (b) Maximum resistance at zero magnetic field R_{max}, and saturation resistance R_{sat} as a function of annealing temperature.

distinct change after $T_{an} = 220°C$. Below this temperature there is a small increase in the GMR signal, while above 220°C there is a dramatic reduction in the GMR. The initial increase in GMR is associated with a reduction of overall resistivity (up to 150°C) and changes in the structure that are reflected both in the X-ray and magnetic measurements. These measurements will be discussed later. By $T_{an} = 300°C$ the GMR effect has nearly vanished, and there is also a significant increase in the overall resistivity. This increase in resistivity seems to be associated with the deterioration of the layer structure as found by X-ray and magnetic measurements.

Our results are substantially different from those found in the Co/Cu multilayers. Fig. 2a shows that the breakdown of the GMR occurs in the NiFe/Cu system at lower temperatures than in similar Co/Cu multilayers. Furthermore, the increase in resistivity above $T_{an} = 220°C$ is very different from the situation in Co/Cu multilayers where a continuous reduction of the electrical resistivity with increasing annealing temperature is observed [5,12].

Depth-profile investigations of NiFe (100 nm)/Cu (200 nm) stacks by means of Auger Electron Spectroscopy showed that interdiffusion between the individual layers sets in above 250°C [4], with Ni atoms preferentially diffusing into the Cu layers. On the one hand, the annealing leads to defect reduction and grain growth as indicated by XRD (see below). Both effects contribute to a reduction of the resistivity. On the other hand, above a critical temperature of ~250°C, the increase of the resistivity due to the interdiffusion dominates.

3.2. X-ray analysis

We used X-ray reflectometry (XRR) and XRD to trace the effect of annealing on the structural properties of the multilayers. The reflectometry investigations show Bragg peaks due to interference at the bilayer sequence indicating a stable layer stacking up to $T_{an} = 300°C$, and subsequently the beginning dissolution of the layered structure which completely disappeared at $T_{an} = 600°C$ (Fig. 3). In contrast, the total metallic layer with a thickness of 120 nm remained preserved as indicated by the Kiessig oscillations,

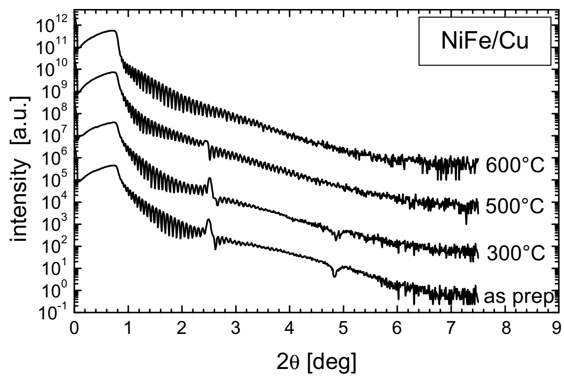

Fig. 3. XRR curves after selected anneals, which indicate the changes of the layer structure.

Table 1
Parameter σ_{rms} of the mean interfacial rms-roughness in nm (typical error: ± 0.05 nm)

	As-prep.	150°C	220°C	300°C	400°C	500°C	600°C
σ_{rms}	0.50	0.50	0.60	0.60	1.10	1.25	—

which remain well pronounced even at $T_{an} = 600°C$. Formally, the variation of the layer structure can be described by an increasing roughness parameter for the NiFe/Cu interfaces (Table 1). In specular reflectometry, however, the influence of an increasing morphological roughness and the onset of interdiffusion cannot be distinguished.

There are several possible reasons why there could be a strong decay in GMR at $T_a = 300$ K and only a weak increase in the roughness parameter. First, the depth-profile investigations [4] shows that Ni, but not Fe, diffuses preferentially into the Cu layers above 250°C. Therefore, even with strong intermixing between Ni and Cu the contrast between Fe-containing and Fe-free layers would be preserved for the XRR measurements. Secondly, the outdiffusion of Ni changes the electronic and magnetic properties of the initial Permalloy towards an Invar alloy possessing a lower magnetic moment. Finally, the afm coupling of adjacent NiFe layers could change (since we have Ni loose spins in the Cu layers [13]), and the effective layer thickness for the changed NiFe composition could be different. The last effect is clearly smaller in the NiFe/Cu multilayers in comparison to the Co/Cu system, where substantial changes of the effective layer thickness during annealing have been observed [5].

We note that a minor interdiffusion already modifies the interface properties significantly. As we will see below, the transport and magnetic properties are sensitive to subtle changes in the NiFe/Cu structures occurring already below 300°C, while the layer and interface characterization by XRR shows substantial structural changes only in the temperature region above 300°C. The same tendency was observed by off-specular XRR measurements which indicate a strong roughness correlation between the interfaces of adjacent layers [10]. Significant changes in the diffuse scattering of the NiFe/Cu multilayers could be resolved only above 300°C.

The wide-angle diffraction patterns (not shown here) indicate a predominating $\langle 1\,1\,1 \rangle$ texture consisting of small grains with a typical size in vertical direction, D_{vert}, of about 25 nm for the as-deposited state. Typically the films show a columnar growth with lateral extensions of the grains smaller than D_{vert}. Due to the annealing, grain growth sets in between 220°C and 300°C. Up to $T_{an} = 500°C$ the vertical grain size D_{vert} increases to about 50 nm. Since other contributions for line broadening such as interface roughness and lattice defect contributions could not be separated, this value of D_{vert} should be considered a lower limit; it seems reasonable that a certain fraction of crystallites grew through the complete layer stack, as has also been observed in Co/Cu multilayers. Additional grain orientation measurements by SEM using the electron back scattering diffraction (EBSD) technique were performed on annealed samples. For the as-deposited state, this technique fails due to the too small grain size of about 20 nm. After the 400°C anneal EBSD patterns were obtained indicating a pronounced fraction of crystallites of about 300–500 nm lateral extension and a mean lateral size of observed grains of about 600 nm. Though the layer stack still contains crystallites with a size below 100 nm which could not be identified by EBSD, some grains grew up to 1 μm lateral extension and larger. Interestingly, the annealing at 600°C leads not to a larger lateral grain size, but rather to a more uniform grain growth with narrower size distribution, i.e. the fraction of crystallites below 100 nm and above 600 nm decreased (Fig. 4).

The ratio of the fraction of crystallites with $\{1\,0\,0\}$ planes parallel to the surface and those with the $\{1\,1\,1\}$ planes parallel to the surface remained constant at about 0.1; this ratio is obviously unaffected by the grain growth introduced by higher annealing temperatures. In contrast to this observation, a texture alteration in comparable Co/Cu GMR multilayers occurs for the same annealing conditions [12]. Apparently, the different changes at the interfaces of both systems correlate with the different development of the

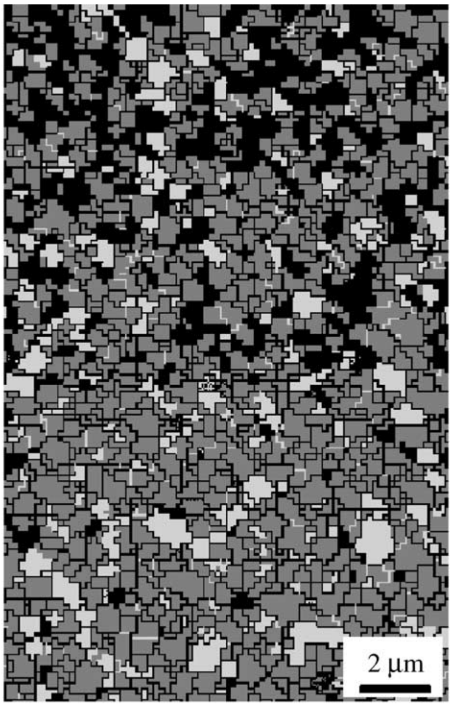

Fig. 4. Orientation mapping of the NiFe/Cu multilayer after annealing at 600°C. Grain areas are light grey for ⟨1 1 1⟩ grains, dark grey for ⟨1 0 0⟩ grains and black for grain boundaries and areas where no indexing was possible.

texture during annealing. Whereas in the Co/Cu system strong satellite reflections appear indicating coherency strains, the onset of interdiffusion prevents this in the NiFe/Cu system. Nevertheless, the sharpness of the ⟨1 1 1⟩ texture continuously increased as we increased the annealing temperature. To characterize the ⟨1 1 1⟩ texture, we measured cuts through the ⟨1 1 1⟩ pole figure as shown in Fig. 5. In the as-prepared state, the texture was relatively broad with a half-width $FWHM_\psi$ of 21°, which decreased to 12° after annealing at $T_{an} = 500°C$.

Apart from the texture differences between the Co/Cu and NiFe/Cu multilayers, the final structure after the complete multilayer decomposition (reached for the NiFe/Cu sample at 600°C according to XRR) is also completely different in both systems. In Co/Cu multilayers, the common multilayer diffraction peak splits after annealing at 740°C into two separated peaks, which are

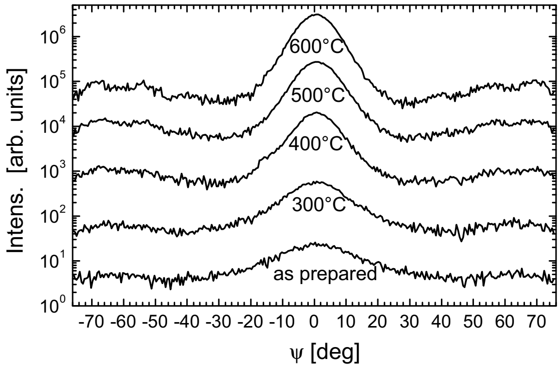

Fig. 5. Cuts through the ⟨1 1 1⟩ pole figure, which indicated the texture sharpening.

attributed to the individual components [8]. This corresponds to the formation of large domains of pure Co and Cu, each of which is scattering coherently. In contrast, the NiFe/Cu multilayer peak develops without significant changes into a single broad peak of the final NiFeCu alloy without detectable signs of single-component domains.

Additionally, the mechanical stress in the layers changed as the annealing temperature was increased. In the as-deposited state compressive stresses of about –100 MPa were evaluated by the $\sin^2\psi$-method from measurements of the {1 1 1} reflection in the Ψ-geometry [10]. Due to the annealing, increasing tensile stresses developed caused by the higher thermal expansion coefficient of the multilayer as compared to the substrate. After the 600°C anneal a mean biaxial stress of 650 MPa developed in the layer stack, presumably supporting the formation of lattice defects and the degradation of the multilayer structure.

3.3. Magnetic measurements

We used MOKE and FMR techniques to determine the magnetic properties of the NiFe/Cu multilayers. Our MOKE measurements were carried out for various rotational angles around the normal to the sample surface. Along one axis "S"-shaped loops were obtained (this turned out to be the hard axis), and along the axis 90° off the original one we observed hysteresis loops such as in Fig. 6a, which are typical for antiferromagnetically coupled systems. The presence of both biquadratic and bilinear coupling contributions is demonstrated by two critical fields, H_1 and H_2, where H_1 represents the initial deviation from the co-linear (saturated) configuration and H_2 represents a field at which the neighbouring layers become antiparallel. Note that this shape of the hysteresis loop, characteristic for biquadratic coupling, is only visible if the easy axis of the magnetization is parallel to the applied magnetic field, a condition which was not fulfilled for the GMR measurements. The annealing of the multilayers below 260°C did not reveal any changes in H_1 and H_2. The angular measurements and a numerical simulation allowed us to determine not only the values of the exchange coupling, but also the strength of the small uniaxial in-plane anisotropy ($H_u = 20\,\mathrm{Oe}$). The strength of the exchange coupling ($J_1 = -0.0012\,\mathrm{erg/cm^2}$ and $J_2 = 0.001\,\mathrm{erg/cm^2}$) was very small and nearly constant up to an annealing temperature of 260°C. At this temperature the hysteresis loops showed visible deformations with respect to the as-grown data—see Fig. 6b.

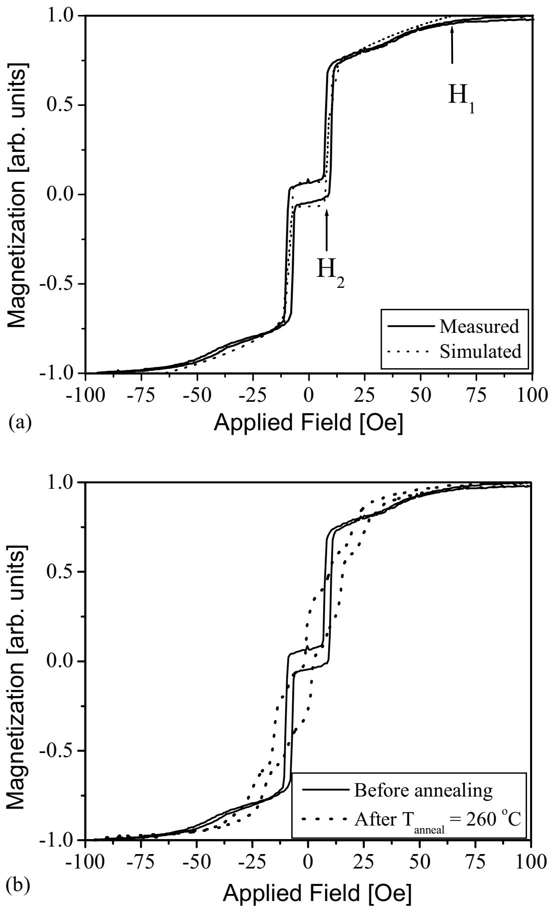

Fig. 6. (a) The hysteresis loop measured along the easy axis (solid line) and the simulated curve (dotted line) The hysteresis loops of NiFe/Cu structures measured after deposition (solid line) and after annealing at 260°C.

The FMR measurements confirmed the presence of the uniaxial in-plane anisotropy. Fig. 7 shows the FMR resonance field after annealing at 180°C as a function of the angle. The values of the uniaxial in-plane anisotropy determined from FMR measurements were similar to those determined by the fitting of the MOKE data [14]. The uniaxial in-plane anisotropy ($20 \pm 5\,\mathrm{Oe}$) was nearly constant up to an annealing temperature of 260°C. Then we observed a significant increase to 60 Oe at 330°C. The $4\pi M_{\mathrm{eff}}$ behaved in similar fashion. Up to 260°C, the value of $4\pi M_{\mathrm{eff}}$ was nearly constant at 6.5 kG, followed by a rapid decrease to 5.6 kG at 330°C.

The measurements of the FMR linewidth revealed an interesting behaviour (Fig. 8). The linewidth was nearly constant ($\Delta H = 120\,\mathrm{Oe}$) up

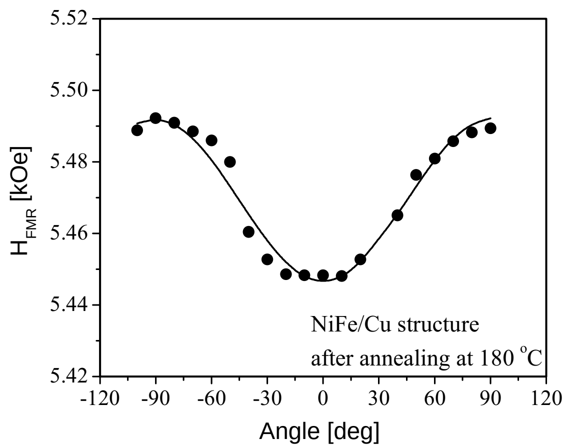

Fig. 7. Angular dependence of the FMR resonance field after annealing at 180°C.

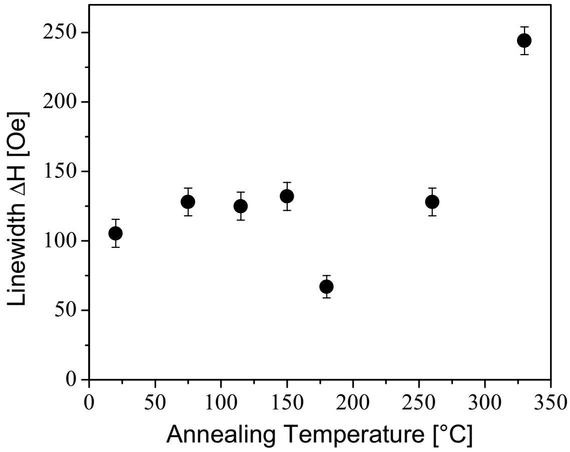

Fig. 8. FMR linewidth as a function of annealing temperature, measured at 24 GHz.

to an annealing temperature of 150°C. After annealing at 180°C the linewidth decreased to 75 Oe. Interestingly, this minimum of the FMR linewidth appears just below the temperature for the GMR breakdown where the GMR was maximum. If the linewidth is taken as a measure of the homogeneity of the magnetization within the NiFe layers [15], this could explain the observed GMR maximum. After annealing at higher temperatures we observed again an increase of the linewidth, which correlates to the onset of layer intermixing and degradation of the multilayer structure.

All the above-mentioned observations were made on the acoustic mode of the FMR signal measured at 24 GHz. We did not observe an optical mode during our investigations, however, this is not surprising since the value of the afm coupling was very small and all the layers had nominally identical thickness [16].

4. Conclusions

The combination of the structural, transport and magnetic techniques allowed us to obtain a clear picture of the influence of annealing on physical properties and structure of NiFe/Cu multilayers. All measurements indicated that annealing temperatures above 250°C significantly alter the physical properties of the NiFe/Cu multilayers. In particular, two tendencies can be inferred from the structural investigations. For annealing temperatures below 250°C, the increase of the grain size and the reduction of defects resulted in a narrowing of the FMR linewidth. Such a narrowing of the FMR linewidths indicated that the magnetic homogeneity of the layers improved near the annealing temperature of 200°C, and, in combination with the defect reduction, resulted in a small increase of the GMR effect. For annealing at temperatures above 250°C, we observed significant intermixing between Ni and Cu atoms not only by using X-ray techniques, but also by AES depth-profile studies. Such intermixing resulted in the decrease of magnetic homogenity for the NiFe/Cu structures and we observed a significant decay of the transport properties (GMR) and correlated these changes with the magnetic properties (FMR linewidth increased significantly and $4\pi M_{\mathrm{eff}}$ decreased). In conclusion, it is the alloying tendency of Ni and Cu above 250°C that determines the decay of the GMR and the change in the magnetic properties of FeNi/Cu multilayers.

Acknowledgements

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