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Mismatch-induced recrystallization of giant magneto-resistance (GMR) multilayer systems

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Abstract

The giant magneto-resistance (GMR) multilayer systems NiFe/Cu and Co/Cu have been studied regarding thermally induced recrystallization. The microstructural reaction takes place during brief annealing at 350–450 °C and is accompanied by a change in the crystallographic orientation from ⟨1 1 1⟩ to ⟨1 0 0⟩ wire texture. The reaction may be utilized to produce GMR sensor layers of remarkable thermal stability. It is shown that the crystallographic reorientation is triggered by the minimization of lattice mismatch elastic energy. Although the systems of interest are equivalent in respect of the observed phenomenon, the $\mathrm{Ni}_x\mathrm{Fe}_{(1-x)}$/Cu system is chosen for a detailed analysis because it allows for precise control of the lattice constant by varying the Fe content in the $\mathrm{Ni}_x\mathrm{Fe}_{(1-x)}$ layer. Moreover, the degree of lattice mismatch exerts a critical influence on the recrystallization probability.

Keywords: Giant magneto-resistance (GMR); Thin films; Recrystallization; Lattice mismatch; Elastic energy

1. Introduction

The discovery of antiferromagnetic coupling (AFC) in multilayer film structures consisting of alternating ferromagnetic and non-ferromagnetic materials [1] and the giant magneto-resistance (GMR) effect found shortly afterwards [2,3] triggered a huge amount of fundamental research and considerable industrial interest. Due to their better sensitivity, GMR reading heads made possible recording densities beyond 200 Gbit/in.^{2} (GMR read heads in combination with perpendicular recording), a performance that was inconceivable with older thin film inductive or even magneto-resistive (MR) reading heads. Nowadays, the magnetic medium itself, rather than the reading head, represents the limiting factor due to the superparamagnetic effect. Owing to further advantages, such as large output signal level, low power consumption, small size and relatively low costs, GMR sensing devices have found a growing range of new applications.

Short-term thermal stability at temperatures in the range of 200–400 °C during manufacturing, and for some applications the long-term stability beyond the conventional 100–150 °C, is a crucial issue, e.g. orientation and rotation speed sensors in combustion engines, for which the automotive industry has an urgent demand. Experiments carried out on Co/Cu and Py/Cu systems ('Py' is the abbreviation used here for permalloy – a $\mathrm{Ni}_{81}\mathrm{Fe}_{19}$ alloy) revealed the degradation of the GMR effect amplitude at about 150 °C and 120 °C, respectively [4,5], if the thickness of individual layers was optimized for the first AFC maximum. Systems optimized for the second AFC maximum, which are often used for practical purposes due to their better reproducibility and stability, lose their GMR properties at about 400 °C and 250 °C, respectively [6,7].

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Rather different mechanisms have been suggested to explain the GMR degradation and underlying microstructural changes. In general, Co/Cu and Py/Cu systems may be expected to behave quite differently due to the rather different thermodynamics of both systems. Transmission electron microscopy (TEM) studies of Co/Cu multilayer samples performed by Rätzke et al. [8] showed the transport of Cu into the Co layers along the grain boundaries. If the width of resulting local breaks of the Co layers corresponds to the thickness of AFC, it may affect the GMR by interference effects. Following Monte-Carlo simulations and experimental observations performed by Bobeth et al. [9], formation of pinholes in Co layers with subsequent fusion of these layers by coalescence of resulting bulges on the edges of holes could be responsible for the GMR breakdown caused by ferromagnetic shortcuts. For the case of Py/Cu, grain boundary diffusion of Ni into the Cu layer was initially proposed [10] to form ferromagnetic bridges between Py layers. However, recent 3DAP analysis [11] at relevant temperatures has demonstrated only minor grain boundary diffusion and identified a slight interdiffusion at the interfaces due to non-equilibrium point defects as the important mechanism.

2. Recrystallization and texture transformation

In addition to atomic transport by either volume or grain boundary diffusion, a further interesting process causes a dramatic modification of the microstructure of these systems. As proven by TEM and wide angle X-ray diffractometry, Co/Cu, Py/Cu/Co/Cu and Py/Cu multilayer systems reveal in the as-prepared state a columnar grain structure with a well-defined $\langle 1\,1\,1\rangle$ wire texture. The columnar grains with an average lateral size of 10–20 nm spread in the normal direction across several single layers, as shown in Fig. 1a, c and e. During a thermal treatment at 350–500 °C, a recrystallization takes place, which leads to grain growth by orders of magnitude (see Fig. 1b, d and f), so that the layers end up in a quasi-single-crystalline structure. Moreover, the texture reorients into a $\langle 1\,0\,0\rangle$ direction perpendicular to the layers as is demonstrated by the respective diffraction patterns shown in the inserts of Fig. 1a–d. This recrystallization is also clearly demonstrated by wide angle X-ray diffractometry. Such X-ray patterns obtained in Bragg–Brentano geometry before and after the thermal treatment are shown in Fig. 1g and h for Py/Cu.

As indicated by the TEM micrographs in Fig. 1 and proven by recent 3DAP investigations [11], the chemical integrity of the layer structure is to a great extent preserved or even enhanced by the thermal treatment. Furthermore, the improvement of reflection satellites, which are to be expected for coherent multilayer systems with single film thicknesses thinner than some 10 nm [12], confirms the enhancement of the layer structure (compare Fig. 1g and h). This finding is rather surprising, at least in the case of Py and Cu layers, since Ni and Cu are completely miscible above 354.5 °C.

Since the RKKY interaction is controlled by the Fermi surface of the paramagnetic spacer, the thickness of maximum AFC depends on the crystallographic orientation of the Cu layers. Thus, the observed texture reorientation should influence the magneto-resistivity of a given system. In Fig. 2 the change in GMR amplitudes by a heat treatment of 1 h at 500 °C is shown for different spacer layer thicknesses. The figure shows almost complete degradation of the GMR amplitude for a Cu layer thickness of 2.05, corresponding to 2nd AFC maximum in $\langle 1\,1\,1\rangle$ orientation. However, for a Cu layer with a thickness of 2.77 nm, corresponding to the 2nd AFCM in $\langle 1\,0\,0\rangle$ orientation, an increase in the GMR amplitude is observed. Furthermore, a specimen prepared with a $\langle 1\,0\,0\rangle$ texture via a suitable pre-annealing remains stable during subsequent long-term heat treatments at rather high temperatures (e.g. 400 °C for 64 h). Thus, the recrystallization offers the possibility of producing sensor layers of very high thermal stability.

In consequence, it is expected that the described recrystallization will affect significantly the GMR amplitude of a layer system. The understanding of this recrystallization phenomenon is a prerequisite for controlling GMR degradation at high temperatures. In the following, we will focus on the driving force of the described transformation. In general, either the anisotropic interfacial energy or anisotropic elasticity can be the determining factors. With our experiments the actual driving force will be identified by controlling the lattice mismatch and thus the elastic energy. Experimental findings are interpreted by theoretical estimations of the contributions to the driving force.

3. Experimental procedures

The GMR multilayers studied in the current report were prepared in different laboratories using ion beam and magnetron sputtering techniques. Thermally oxidized silicon wafers of [1 0 0] orientation and an oxide thickness in the range of 50 nm to 1.5 μm were used as substrates. As measured by atomic force microscopy (AFM), the root mean square (RMS) roughness amounts to 0.3 nm. In some cases conventional glass substrates were used as well.

The main series of $Ni_xFe_{(1-x)}$/Cu multilayer systems, which are of major importance for the present study, were produced in the laboratory at Münster University. An ion beam, ultrahigh-vacuum (UHV) system, similar to that described in Ref. [13], was employed. A quartz crystal thickness monitor guaranteed a reproducible thickness of the deposited structures. A pure Cu (99.9%) target and a series of $Ni_xFe_{(1-x)}$ targets with different iron contents (0–70 at.%) were used. The $Ni_xFe_{(1-x)}$ targets were melted in an arc furnace from pure Ni (99.98%) and Fe (99.9%). The typical working pressure of argon gas, ion beam voltage and current during deposition were 2×10^{-2} Pa, 800 V and 35 mA, respectively. Residual gas pressure was better than 1×10^{-5} Pa. The sputtering rates of $Ni_xFe_{(1-x)}$ and Cu were set to be 2.1–2.3 Å/s and 3.7 Å/s, respectively. Before deposition, the substrates were chemically cleaned