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Acta Materialia 53 (2005) 3383–3393

Acta MATERIALIA

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Stability and thermal reaction of GMR NiFe/Cu thin films

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Received 16 October 2004; received in revised form 23 March 2005; accepted 29 March 2005
Available online 13 May 2005

Abstract

Giant magneto-resistance model systems of NiFe/Cu multilayer stacks with 2 nm single layer thickness were deposited onto needle-shaped W tips using ion beam sputtering and analyzed by atom probe tomography after appropriate heat treatments. Owing to the outstanding sensitivity of the method, even minor chemical modifications on the nanometer scale can be detected. Although annealing treatments at temperatures up to 250 °C already result in a dramatic decrease of magneto-resistivity, no major structural or chemical transformation of the initial layer system is found. Instead, a slight decrease of the concentration slope at the interfaces is observed, which is attributed to short range interdiffusion induced by non-equilibrium point defects. Annealing at higher temperatures up to 500 °C/40 min still preserves a clear layer structure. However, appreciable amounts of Ni are dissolved inside the Cu layers. In the presence of grain boundaries, the onset of significant grain boundary diffusion occurs at about 350 °C.

Keywords: Magnetic multilayers; Giant magneto-resistance; Diffusion; Three-dimensional atom probe

1. Introduction

In recent years reading heads based on the giant magneto-resistance effect (GMR) have made possible a dramatic increase in magnetic recording density. Even more than a decade after the discovery of the effect in Fe/Cr thin film multilayers by Baibich et al. [1] and Binash et al. [2], magnetic multilayers still attract a remarkable amount of scientific interest because of their already proved utility in data storage and magnetic sensor technique. Co/Cu and Cu/$Ni_{79}Fe_{21}$ (permalloy = Py) are two of the most often used systems. Co/Cu presents higher GMR amplitudes, but a pronounced hysteresis reduces the sensitivity and introduces some ambiguity in the signal of positional or orientational sensors. In this case, Py/Cu multilayers are promising candidates because of their higher sensitivity and lower hysteresis.

However, for many potential applications, as e.g., angular sensors in motor vehicles, the thermal stability of the device is an important issue. It is known that the Cu/Py system is much more sensitive to thermal load than Cu/Co. Heat treatments at a temperature of 150 °C already modify the GMR amplitude of Cu/Py multilayers and above 250 °C the GMR amplitude disappears almost completely [3–5]. By contrast, the GMR amplitude of Cu/Co systems remains unchanged up to 400 °C [5]. Different explanations for the mechanisms, which are responsible for the GMR degradation, have been proposed: van Loyen et al. [3] argue that at least two different mechanisms contribute to the breakdown of Cu-based GMR multilayers (Co/Cu and Py/Cu), namely grain boundary diffusion and inter- or de-mixing at the interface. Grain boundary diffusion of Ni in Cu was found also by Schleiwies et al. [6] in a Co/Cu/Py

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doi:10.1016/j.actamat.2005.03.038

triple layer using local chemical analysis. After a complex investigation of a $Py_{100\ nm}/Cu_{200\ nm}/Py_{100\ nm}$ model trilayer, including resistance measurements, Auger electron spectroscopy, X-ray diffraction and laser-optical stress measurements, Brückner et al. [7] conclude that interdiffusion over only short distances dominate the degradation of the GMR amplitude in Cu/NiFe multilayers at moderate temperatures. Hecker et al. [5] have also investigated the effect of annealing on a Py/Cu multilayer by X-ray diffraction, electron microscopy, measurements of transport properties, magneto-optical Kerr effect or ferromagnetic resonance and they conclude that the alloying tendency of Ni and Cu above 250 °C determines the decay of the GMR and the change in the magnetic properties of the NiFe/Cu multilayers.

The remarkable difference between the thermal stability of Co- and Py- based systems is due to the thermodynamics of the respective phase diagrams. While Cu and Co are nearly immiscible, Cu and Ni form a continuous range of solid solutions with a solubility gap determined rather poorly at low temperatures. Thus, the latter system is already unstable in terms of bulk thermodynamics, whereas the first one may be only destabilized by interfacial contributions. However, we are interested in the reaction mechanisms in detail: which elements are the mobile ones, at which point is the structure of dense layers destroyed, what is the role of structural defects like grain boundaries or interfacial roughness? Since the interesting periodicity of the multilayers is restricted by the Ruderman and Kittel, Kasuya and Yosida (RKKY) coupling to the range of 2–4 nm, state of the art analysis methods of highest spatial resolution are required. A further complicating factor arises, if the clear layer structure is disturbed in later stages of the reaction, so that three-dimensional information is needed to understand phenomena like the transport along the grain boundaries or the formation of local layer breakthroughs. Therefore, in this study atom probe tomography (3DAP) is applied; a technique which allows a three-dimensional reconstruction of the spatial distribution of the various atomic species with single atom sensitivity and a spatial resolution in the range of 2–4 Å slightly depending on the analysis direction.

The 3DAP technique has been used successfully over the last few years to study magnetic multilayers. Investigations were conducted by Larson et al. [8,9] on different systems exhibiting GMR. In a $(Ni_{82}Fe_{18}/Co_{90}Fe_{10}/Cu/Co_{90}Fe_{10}) \times 10/Ni_{82}Fe_{18}$ grown on Si, they found that there is an enrichment of Fe at the interface were Cu is grown on CoFe and a depletion of Fe where CoFe is grown on Cu [8]. Investigating $Cu_{2\ nm}/Co_{2\ nm}$ annealed at 360 °C for 1 h, they found an increase in grain size, but no apparent change in the interfacial mixing between Cu and Co. They studied also CoFe/Cu films deposited in the presence of oxygen and observed that the GMR of these specimens becomes more sensitive to heat treatment compared to layers produced in a clean vacuum atmosphere. No detailed physical mechanism could be postulated for this effect yet, which may be partly due to the fact that they studied only very few specimen states owing to their fascinating but complicated preparation method [9].

2. Experimental details

The specimen preparation for atom probe tomography requires particular effort, especially in the case of thin film multilayer structures. Only a few techniques may be applied to position the interfaces of interest into a needle-shaped tip of a radius of curvature below 50 nm. Tips can be cut out of planar layer systems by lithography techniques [10] or by deposition on Si posts and subsequent shaping by means of a focussed ion beam [11]. However, these methods require several, rather difficult and time consuming steps, so that the study of a sequence of reaction stages becomes almost impossible in practice.

Therefore, for the present study layer systems were directly deposited onto pre-shaped tungsten tips using a dedicated ion beam sputtering system [12]. Using a conventional field ion microscope, tungsten substrates were field developed up to 12 kV before coating to obtain a smooth surface and suitable radius of curvature. In order to get stable specimens, remaining adsorbants on the substrate surface were removed by Ar ion etching directly before deposition. $Py_{5\ nm}/(Cu_{2.5\ nm}/Py_{2.5\ nm})_{3-5}/Cu_{10\ nm}$ multilayer stacks were deposited by ion beam sputtering from a pure Cu and an alloy target of the ideal composition ($Ni_{79}Fe_{21}$). Deposition was carried out at room temperature using a base pressure of 5×10^{-6} Pa, an Ar pressure of 1×10^{-2} Pa and a growth rate of 3–5 nm/min. We have shown in a previous study that the curved substrate induces in deposited metallic thin films a smaller grain size compared to layers on flat substrates. However, typically a single larger grain develops on the (0 1 1) pole of the tungsten surface close to the tip axis, so that in the majority of measurements no grain boundary is found in this region [13].

Appropriate heat treatments were carried out in a vacuum furnace of 1×10^{-6} Pa base pressure. The analysis was performed with the tomographic atom probe at the University of Göttingen, Germany [14], which was built around a detector system developed at the University of Rouen [15]. Suitable measurement conditions were found for a pulse rate of 2 kHz, a pulse fraction $U_{dc}/U_{pulse} = 25\%$ and a specimen temperature of 40 K. To determine the effect of volume diffusion, which is expected to proceed homogeneously along the interfaces, measurements were performed in random area mode, i.e., tips are oriented approximately parallel to the flight