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Thermal stability of a Co/Cu giant magnetoresistance (GMR) multilayer system

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ABSTRACT

Critical limitations exist regarding the thermal stability of multilayer giant magnetoresistance (GMR) sensors due to significant contribution of layer interfaces and grain boundaries to their performance. This paper addresses the stability of, and thermal reaction in Co/Cu magnetoresistive systems. Thin film multilayers were deposited by ion beam sputtering on top of preformed tungsten tips. The multilayers were then analyzed by a wide-angle tomographic atom probe (WATAP) in the as-prepared state and after heat treatments in the temperature range from 150 to 550 °C.

As revealed by the WATAP analysis, Co breakthroughs along grain boundaries in the Cu spacers appear at 450 °C. This morphological change leads to a deterioration of the GMR effect due to the magnetostatic ferromagnetic coupling between Co layers. Furthermore, a strong effect of Cu layers faceting is observed after annealing in the same temperature range, which may lead to the deterioration of the layered structure on high-angle grain boundaries.

1. Introduction

Since the discovery of antiferromagnetic coupling (AFC) [1] and giant magnetoresistance (GMR) effect [2,3], a huge amount of fundamental research have been undertaken in order to increase the magnetoresistance (MR) ratio, sensitivity, and thermal stability of the GMR materials. Owing to significant advantages over the predecessor MR sensor technologies, the areas of GMR sensor applications have expanded continuously. Especially in the automotive industry, there is a great demand in high sensitivity, small size, and low cost magnetic field sensors. Linear and rotary position sensors, gradiometers, ignition timing sensors, and sensors for measuring current, acceleration, and pressure give a full control over a car functionality. Thermal stability of such sensors is a crucial issue for the applications in the combustion engine environment (temperature range up to 400 °C).

The very small size of state-of-the-art magnetic sensors based on the GMR effect, and significant contribution of interfaces and grain boundaries to their performance, makes these devices inherently unstable. Experiments carried out on Co/Cu GMR multilayers revealed degradation of the GMR effect amplitude at about 150 °C [4], 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 [5].

Rather different mechanisms have been suggested to explain the GMR degradation and underlying microstructural changes in the Co/Cu multilayers. Observations of Co bridges through Cu layers have been reported by Larson et al. [6] and Langer et al. [7] by means of field ion microscopy (FIM) and transmission electron microscopy (TEM), respectively. These defects of the layered structure were observed in systems with high roughness of layer interfaces even in the as-prepared state. Evidently, this effect leads to strong ferromagnetic (FM) coupling of adjacent Co layers. TEM studies of Co/Cu multilayer samples performed by Rätzke et al. [8] show 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 amplitude by frustration effects. From 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 can be responsible for the GMR breakdown caused by FM shortcuts.

In order to shed the light onto the annealing-induced chemical and microstructural changes taking place in the Co/Cu multilayers, atom probe tomography (APT) was applied in this study. This technique yields real 3D information with a spatial resolution of a few angstroms and furthermore single-atom sensitivity. It allows for direct observations of the 3D atomic species configurations on the subnanometer scale.

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2. Specimen preparation

The GMR multilayers were prepared using an ion beam sputtering deposition system at Münster University. Atom probe analysis of thin films requires particular efforts in preparation of specimens. The investigated multilayers were deposited on preformed tungsten substrate tips with apex radii of curvature of 30–50 nm. The preparation of FIM/TAP specimens involved:

- electropolishing of tungsten wire;
- field development of the W-tip in a FIM;
- deposition of desired materials;
- heat treatment of the specimen.

The electropolishing was carried out in two molar sodium hydroxide (NaOH) solution by short pulses of 2–7 V of AC voltage in order to produce specimens having an apex curvature radius of 10 nm only. After electropolishing, the tips were developed by means of FIM to form a smooth surface and to increase the radius of the tip apex curvature to some 30–50 nm. The tips were typically developed at 30 K to voltages of about 12–14 kV. Subsequently, the tips were transferred into a sputtering chamber for the deposition of desired materials. An ultra-high vacuum ion beam deposition system, similar to that described in [10], was employed for this purpose. To ensure thickness reproducibility of the deposited structures, a quartz crystal thickness monitor was used. Pure Cu and Co targets with purities of 99.9% and 99.99%, respectively, were used for the depositions. Typical working pressure of Ar-gas, ion beam voltage, and current during deposition were 2×10^{-2} Pa, 550 V, and 15 mA, respectively. The residual gas pressure was better than 1×10^{-5} Pa. Sputtering rates of Co and Cu were 0.4 and 0.75 Å/s, respectively, as monitored using the quartz crystal microbalance. During the deposition, the substrates were maintained at room temperature using water cooling. The last step in the specimen preparation was a thermal treatment in a UHV furnace to produce a desired stage of the material reaction. Thermal treatments were performed directly after deposition without breaking the vacuum, using a ceramic heating plate.

Electrical grounding of the sample during its handling was proven to be a crucial point to avoid formation of amorphous W oxide layer, which deteriorates the quality of the substrate tip and dramatically decreases the success rate of the wide-angle tomographic atom probe (WATAP) analysis (for further information see Refs. [11,12]).

3. Experimental results

3.1. FIM analysis

FIM analysis was carried out in order to reveal the typical microstructure of the multilayers. It was important to confirm that the microstructure of the multilayers evolved on the special curved tip surface is comparable to those found in multilayers grown on planar substrates. Since the effective field of view in FIM measurements is larger than in atom probe measurements (due to the shorter specimen-screen/detector distance in our FIM compared to WATAP), the information deduced from the FIM micrographs can also be very useful for WATAP data evaluation.

FIM micrographs of the Co/Cu multilayer in the as-prepared state are shown in Fig. 1. The micrographs are obtained by subsequent field evaporation of surface material. The current specimen surface position is indicated in the sketch. As seen from Fig. 1a, the layer stack grows with [1 1 1] orientation along the tip axis. This feature can be deduced from the three-fold symmetry of the observed configuration of the crystallographic poles in the uppermost Cu layer. The FIM micrographs in Fig. 1a and b reveal that the [1 1 1] crystallographic pole of Co is placed exactly under the [1 1 1] Cu pole. The same is observed for ⟨2 2 0⟩ poles of Cu and Co layers (compare Fig. 1a and c). These observations suggest the coherency of the Co/Cu interface structure. Often, a single large grain is detected on the tip apex, which is the typical morphological feature observed in thin film TAP specimens deposited on top of the tungsten tips. Grain boundaries, detected mainly on the tip periphery, propagate through the layer stack down to the substrate, as observed upon subsequent field evaporation of all deposited layers. This implies a columnar grain morphology (this feature is clearly seen in TAP measurements described below). Since no 6-fold symmetry was detected in the FIM micrographs of Co layers, face centered cubic (fcc) structure of Co lattice and not the hexagonal close packed (hcp) structure as in bulk is assumed. It agrees with the previous reports concerning the lattice structure of thin Co films on fcc substrates (see Refs. [13–15]). All the microstructural features described above are similar to those observed in planar specimens (see Ref. [16]). Thus, no significant discrepancy is expected in the thermal behavior of planar and curved multilayers.

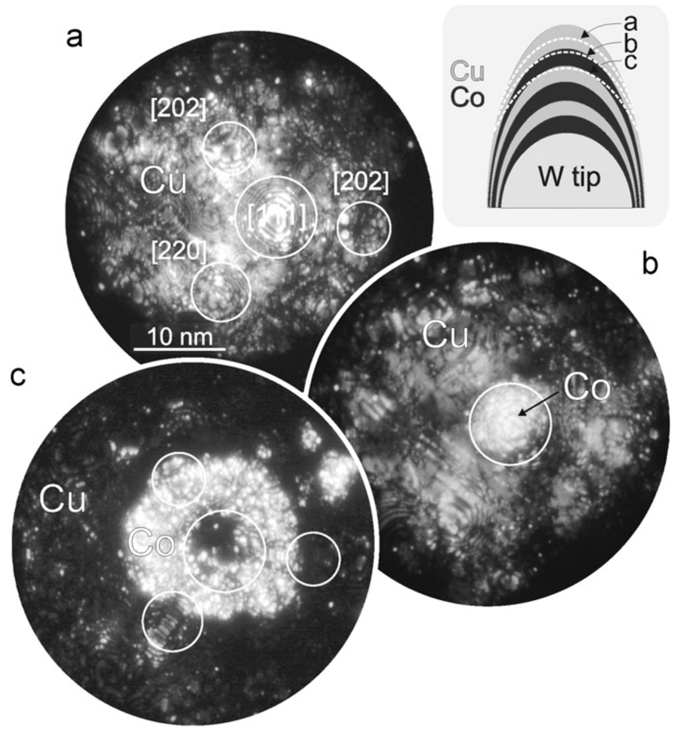

Fig. 1. FIM micrographs of the Co/Cu multilayer in as-prepared state. The micrographs are obtained by subsequent field evaporation of surface material.

3.2. WATAP analysis

In this work, the Co/Cu system was proven to be suitable for atom probe analysis in voltage evaporation mode. It shows sufficient mechanical stability despite a high number of potentially unstable Co/Cu interfaces. It was possible to conduct the measurements at relatively low temperatures (≈ 30 K), yielding measurement data with reasonably low noise levels. Due to good separation of the mass peaks in the mass spectrum, elemental species can be determined without any ambiguity resulting in reliable species identification. The measurements were carried out with a pulse frequency of 20 kHz, a pulse to base voltage ratio