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Nano-analysis of Ta/FeCoB/MgO tunnel magneto resistance structures

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ABSTRACT

The partitioning and segregation of B during the crystallization of amorphous FeCoB in Ta/FeCoB/MgO layered structures is investigated by atom probe tomography to obtain a better understanding of the beneficial impact of Ta capping layers on FeCoB/MgO/FeCoB magnetic tunneling junctions. Boron, initially uniformly dissolved in the amorphous FeCoB layer, is rejected from FeCo grains on crystallization and first segregates at the interface to the Ta layer where it prevents nucleation of crystalline FeCo. Only later, it is fully absorbed by the Ta layer. In the studied thin film structures, B neither segregates at nor dissolves into the MgO barrier even after prolonged heat treatment. Important kinetic parameters are derived from detailed isochronal annealing series. They allow the quantitative modelling of the observed process. The combination of high affinity to B but low diffusivity makes Ta the unique capping material to achieve an optimum performance.

1. Introduction

The theoretical prediction of over 1000% tunneling magnetoresistance (TMR) by the preferential spin-dependent tunneling of certain wave functions in a coherent Fe(001)/MgO(001)/Fe(001) magnetic tunneling junction (MTJ) [1,2] led to the experimental demonstration of giant tunneling magnetoresistance (TMR) above 150% at room temperature in MgO based MTJs [3,4]. Subsequently, Djayaprawara et al. [5] reported that a heat-treated CoFeB/MgO(001)/CoFeB MTJs prepared by magnetron sputtering exhibits a much larger TMR of 230% at room temperature. The high TMR obtained in these MTJs was attributed to the growth of an (001) oriented MgO barrier on an amorphous CoFeB layer, and subsequent templated crystallization of bcc CoFe with grain-to-grain epitaxy between CoFe and the MgO barrier [6,7]. The TMR ratio of CoFeB/MgO/CoFeB MTJ was found to be further improved by deposition of additional capping layers [8–11]. In particular, Ta capping shows a remarkably positive impact on the TMR ratio of the CoFeB/MgO/CoFeB based MTJ, which is currently used in read head sensors in hard disk drives. Since the TMR values are significantly affected by the CoFe/MgO interface and the B segregation after the crystallization of amorphous CoFeB, several attempts have been made to quantify B during the recrystallization process. Miyagima et al. [8] and Karthik et al. [9] investigated the boron distribution in CoFeB/MgO/CoFeB pseudo spin valves by high resolution transmission electron microscopy (HRTEM) and electron energy loss spectroscopy (EELS) with various capping layers. Kozina et al. [12] studied these structures using hard X-ray photoemission HAXPES. They reported a strong dependence of the crystallization process of the CoFeB layer on annealing temperature and capping layer. They stated to see a tendency of boron to diffuse preferentially to the Ta capping layer and rarely to the MgO barrier. Recently, Greer et al. [13] used standing-wave hard X-ray photoemission spectroscopy (SW-HXPS). They deduced a concentration of 19.5% of boron uniformly distributed within the MgO barrier and less amount of boron, up to about 2.5 at%, segregated at the interface to the Ta capping. Although these methods give important insight, the conclusions have been deduced through complex data processing. In general, it is difficult to access the segregation of light elements at buried interfaces without real 3D information in atomic resolution. Therefore, we apply in this work laser-assisted atom probe tomography (LA-APT).

Atom Probe Tomography (APT) is well known as an outstanding analysis technique on the sub-nanometer scale. It delivers three dimensional reconstructions of the atomic arrangement with single atom sensitivity. The recent extension of the atom probe technique

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by laser pulsing extended its application to semi-conductors and even insulating oxide ceramics [14,15]. For a comprehensive description the reader is referred to recent literature [16,17]. APT analysis of functional Fe(Co)/MgO/Fe(Co) TMR sensor films were already performed in recent studies see e.g. Refs. [18–21]. In summary, these studies suggest that the details of the chemical behavior may depend sensitively on the actual deposition conditions and the quality of the produced barriers. Segregation of metallic components at the barrier or even their solution within the barrier have been reported. However, it cannot be excluded that these observations were due to a weak quality of the produced barrier, as it is indicated by inhomogeneous composition laterally along the barrier [18,19] or due to some degree of over-oxidation, so that part of the electrodes became oxidized [20,21]. Here, we focus on the general solid state reactions appearing in such layer structures. Therefore, in order to clearly work out the essential mechanisms, we prepare model structures of partly emphasized thicknesses under ideal sputter conditions. These allow investigating the chemical composition in the barrier and electrodes, the interfaces and the Ta cap in a clearer manner and so to analyze the underlying thermodynamic driving forces and the kinetics of segregation and dissolution of boron in the multi-layered structure of Ta/CoFeB/MgO.

We have already presented a previous study that was limited to model bilayers Ta/CoFeB, which could be analyzed by conventional high-voltage-pulsed APT [22]. In this work, we extend our investigation to the trilayer stack Ta/CoFeB/MgO, which provides a more realistic model to a real TMR device. This is especially important to check a possible solution of B inside the MgO barrier and to understand the epitaxial relation between the barrier and the electrode. In view of the recent success of laser-assisted APT, early attempts of analyzing thin insulating barriers by electrically pulsing, see e.g. Refs. [23,24], need to be seen with some reservation regarding reliability. Here, as a consequence of including thicker MgO layers, the laser-assisted evaporation mode is without alternative to achieve reliable APT data of a statistically significant number of samples.

2. Experimental procedures

Since needle-shaped specimens that were prepared from actual MTJ by the lift-out technique were prone to rupture, we analyzed model Ta/$Fe_{80}Co_{20}B_{20}$/MgO stacks that were deposited on tungsten needles. These substrate needles were produced by electropolishing starting from a 0.075 mm diameter tungsten wire using 2 mol/L NaOH electrolyte. The electropolished tips were field-developed to a well-controlled radius of 30 nm by using a field ion microscope (FIM). Then, triple layers of Ta_{10nm}/$(Fe_{80}Co_{20}B_{20})_{20nm}$/$MgO_{10-20nm}$ were deposited on top of the substrate tips by ion beam sputtering in a custom-made UHV deposition system (residual pressure less than 1×10^{-7} mbar). For comparison, also full TMR stacks were deposited on oxidized Si wafers. The thickness of deposited layers was in-situ controlled by means of a quartz micro balance. In all cases, the oxide barrier was sputter-deposited from a commercial MgO target, without using additional oxygen in the sputtering atmosphere.

Two annealing sequences were carried out in an UHV furnace (residual pressure of less than 10^{-8} mbar): (i) an isochronal series of 1 h annealing at various temperatures ranging from 300 °C up to 800 °C and (ii) an isothermal sequence at 500 °C with durations up to 600 min. Since the atom probe analysis is destructive, an abundant supply of fresh samples had to be prepared. In view of the very small analyzed volume, scatter from sample to sample is unavoidable. A careful averaging on different samples of the same preparation and annealing conditions is required to derive statistically significant results. The annealed structures were analyzed by means of the laser-assisted atom probe tomograph (LA-APT) at University of Münster, Germany [25] using a UV-Laser system ($\lambda = 343$ nm) with a pulse width of 220 fs at 200 kHz pulsing frequency. The typical energy per pulse amounts to about 0.1 μJ. During analysis, samples were cooled to 50 K.

3. Results of the nano analysis

A mass spectrum obtained by APT analysis of a Ta/CoFeB/MgO triple layer is shown in Fig. 1(a). All major peaks can be assigned to the expected elements in various charge states. The two stable boron isotopes are detected as singly or doubly charged ions at 10 u and 11 u respectively 5 u and 5.5 u, while the singly charged ions represent the dominating species. Oxygen and magnesium are observed in different molecular combinations such as Mg^{2+}, MgO^{+1} and the overlapping peaks O_2^{1+}/Mg_2O^{2+}. The latter are deconvoluted according to the natural abundance of the Mg isotopes. Distinguishing the transition metals Fe^{2+} and Co^{2+} as well as Ta^{3+} causes no difficulty. As depicted in Fig. 1(b) (logarithmic scale),

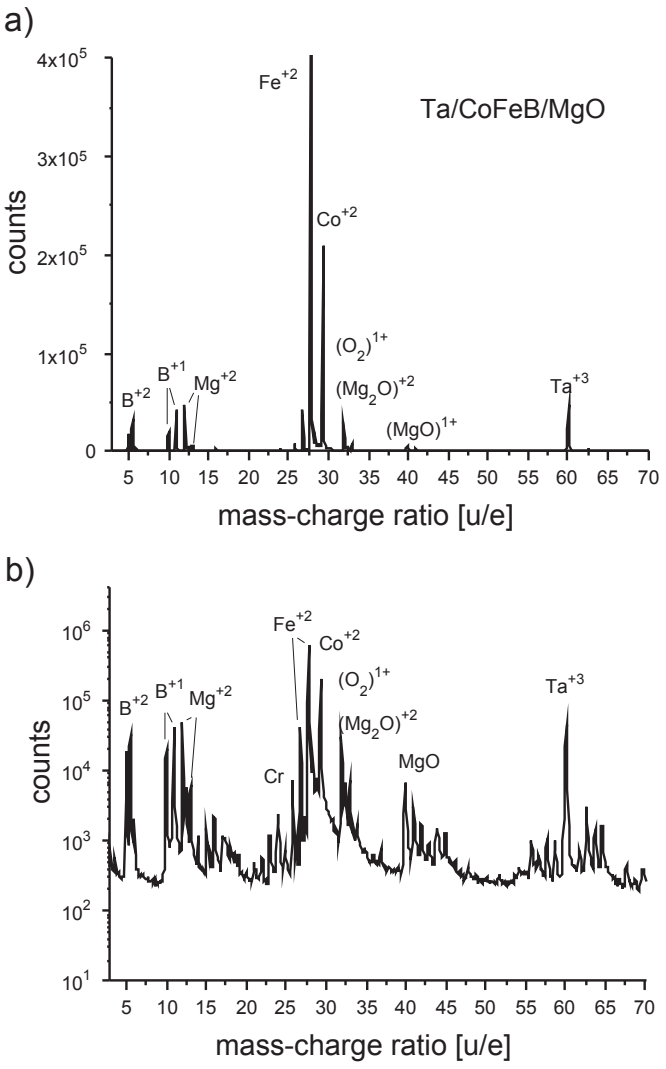

Fig. 1. Spectrum of specific mass (m/z) of a MgO/FeCoB/Ta triple layer as obtained by laser-assisted atom probe tomography, linear scale (a) and logarithmic scale (b). Most components appear well separated. Only the various peaks of Mg and O must be deconvoluted based on the expected abundances of isotopes. Relative levels of impurities is less than 10^{-3}.