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Inter-diffusion of cobalt and silicon through an ultra thin aluminum oxide layer

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ABSTRACT

Optical emission spectroscopy of sputtered species during ion bombardment, Auger electron spectroscopy and high-resolution transmission electron microscopy were used to study the cobalt and silicon diffusion through the interfaces of Co/AlO/Si(0 0 1) hetero-structure. The results are discussed as a function of the annealing temperature of sample and show that the diffusion process at the interfaces starts for annealing temperatures above 200 °C without detectable modification of the oxide layer.

1. Introduction

The magnetic tunnel junction (MTJ) based on ferromagnetic metal/insulator/silicon (MIS) structure is now widely studied for achieving the fabrication of magnetic random access memories (MRAMs) [1,2]. The efficiency of these memories strongly depends on the nature and the physical–chemical properties of the insulating barrier which is usually formed by an oxide layer. Because the spin-polarized electrons transport is performed by tunneling effect, the thickness of the oxide must be very small (~1.5 nm) [2], the composition homogeneous and the interfaces abrupt and smooth at the atomic scale [3].

Moreover, the thermal budget experienced in the integration of the MTJ for their implementation at the "back-end" of CMOS processes may reach 300–400 °C. As well, the normal conditions for MRAMs operation are around 80 °C. Therefore, the thermal stability of MTJ structure in terms of diffusivity, crystallization and interface sharpness is a critical issue. For instance, the diffusion, even at a very low extent of any adjacent element to the oxide layer, can affect dramatically the transport properties of the MTJ [4,5]. The annealing process can strongly reduce the conductance of MTJ because the electronic structure of the oxide/metal interface is affected, as observed for CoFeB/MgO/CoFeB structure [6].

Recently, we have developed a new process to grow aluminum oxide on both silicon and metallic substrates [7,8]. The growth of the oxide layer is performed layer-by-layer following this sequence: (i) evaporation at room temperature (RT) of one aluminum monolayer (ML) on a clean substrate, (ii) RT chemical adsorption of oxygen on the Al ML using low pressure (~2×10^{-6} Torr) of molecular oxygen for 2 min, (iii) annealing at 400 °C for 15 min in Ultra High Vacuum (UHV) conditions (~4×10^{-10} Torr), i.e. after pumping oxygen. This approach, so-called atomic layer deposition and oxidation (ALDO) procedure, is described in more details in references [7,8]. We have shown that the thickness of a single Al/O sequence is ~0.8 nm. Besides, the aluminum oxide obtained following the above process is amorphous with a chemical composition close to AlO, homogeneous throughout the whole thickness of the layer and exhibits a band gap of about 6.6 eV [7,8]. Furthermore, when AlO is grown on a Si(0 0 1)-H terminated surface (i.e. obtained after etching with HF acid dip for the removal of the oxide formed by the last step of a modified Shiraki chemical cleaning) and capped by a thick layer of Co, both Co/AlO and AlO/Si(0 0 1) interfaces are sharp [8,9].

In this paper we present a study of the diffusion barrier properties of this aluminum oxide layer in the case of the Co/AlO/Si (0 0 1)

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hetero-structure using sputter induced photon spectroscopy (SIPS), Auger electron spectroscopy (AES) and High-resolution transmission electron microscopy (HR-TEM).

2. Experimental details

All the samples studied were prepared by the ALDO technique on p-type (10 Ωcm) Si(0 0 1)-H substrates. Aluminum is deposited in the low 10^{-10} Torr pressure range with an evaporation rate of about 1 ML/min. The deposition rate of aluminum (and cobalt) has been calibrated using a quartz balance and AES. The chemical adsorption of oxygen is performed under molecular oxygen pressure of 2×10^{-6} Torr for 2 min, in the same UHV chamber and after isolation of the aluminum source by means of a lock-valve. After each run the formed oxide layer is annealed at 400 °C for 15 min under UHV conditions. Subsequently, a cobalt layer is deposited under UHV pressure to realize Co/Aluminum oxide/Si structures.

The AES experiment is performed in situ in a standard UHV chamber at a 10^{-10} Torr base pressure using electron gun operating at 2.5 keV energy and a RIBER CMA OPC 105 cylindrical mirror analyzer. Auger spectra were acquired in the derivative mode and the data were collected with a computer system allowing an easy measurement of the peak-to-peak height of the Auger signal.

The SIPS technique and its variants are known under several names [10]. This optical spectroscopic technique differs from secondary ion mass spectroscopy (SIMS) with the advantage that the emitted light is not affected by the ionic space charge phenomena. The SIPS experimental set up employed here has been reported in great detail elsewhere [11–13]. Briefly, Kr^+ ions are formed by electron impact on Kr (99.998% purity) in a plasma source. Ion currents of a few micro amperes and of 5 keV Kr^+ beam are transmitted on the target mounted inside an UHV chamber (with a residual pressure lower than 10^{-7} Torr). The ion beam current is measured on the sample by a Faraday cage placed behind the target along the ion beam direction. Typical beam current densities in these experiments were around 1 $\mu A/mm^2$. The angle of the incident ion beam was 60° with respect to the normal of the surface. The sputtering rate is ~1 nm/s and depends on the sample resistivity and the residual pressure while the sensitivity is ~1 ppm [13]. The light emission resulting from atoms extracted from the sample is probed at characteristic wavelengths and therefore the element profile is recorded. The emitted light was analyzed through a HR 320 Jobin–Yvon monochromator equipped with an 1800 groves/mm holographic grating. The light is transmitted onto a Hamamatsu R4220P photomultiplier sensitive in the 190–650 nm wavelength range. During the sputtering, the monitoring of the spectrum light emission allows to follow qualitatively the concentration profile of one element through the interfaces.

HR-TEM experiments were performed with a 300 keV JEOL JEM-3010 microscope having a 0.16 nm resolution. The cross-sectional specimens were prepared by standard mechanical thinning and polishing techniques. Then, they were made electron transparent by low angle and low energy ion milling procedures.

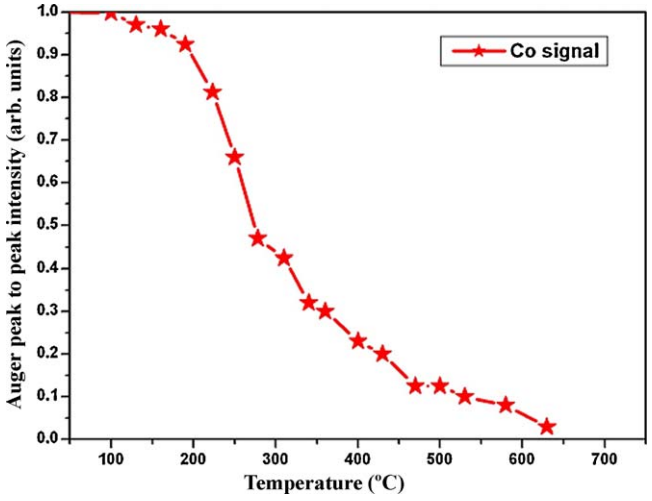

Fig. 1. The Auger peak-to-peak intensity variations recorded during the dissolution of 1 Co ML versus temperature.

3. Results and discussions

The AES measurements were used to access the critical temperature from which the diffusion of cobalt starts in the oxide layer. After deposition of 1 ML of cobalt on AlO layer (4.0 nm thick) previously grown on Si(0 0 1)-H terminated surfaces, we have recorded (see Fig. 1) the variations of the normalized Auger

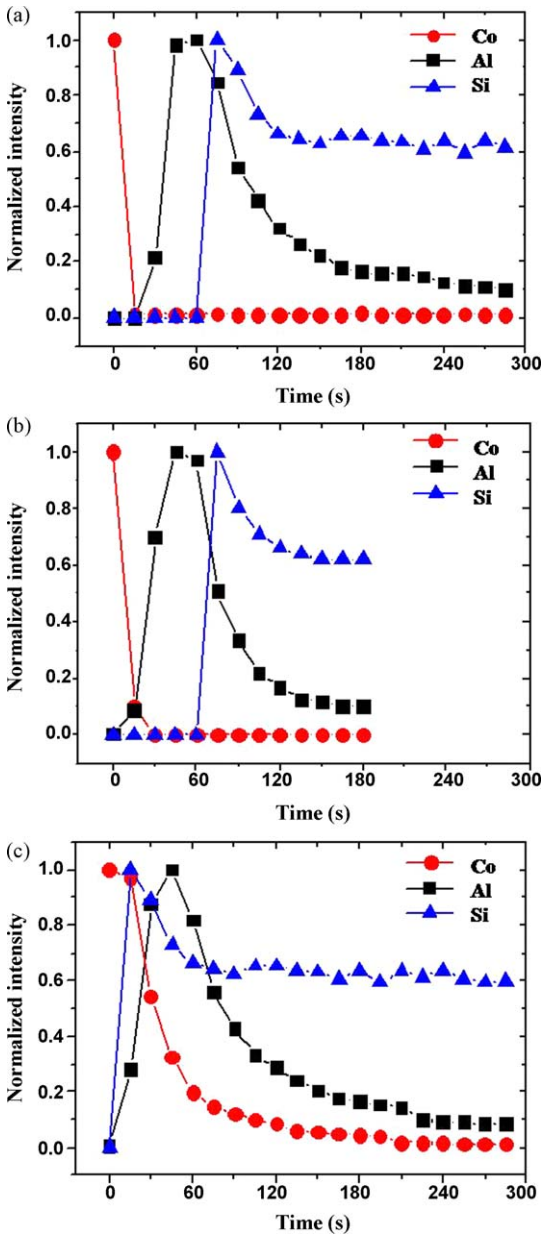

Fig. 2. The SIPS Co, Al and Si profiles corresponding to: (a) as grown sample, (b) sample annealed at 200 °C and (c) sample annealed at 340 °C. All signals have been normalized to their maximum for better presentation.