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Preparation of the subnanometer thick epitaxial $Al_2O_3(0001)$ layers on Fe(110) for magnetic tunnel junctions

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Abstract

Growth as well as crystallographic and electronic properties of thin AlO_x layers on Fe(110) were studied by means of low-energy electron diffraction and Auger-electron spectroscopy. Al layers of different thickness were deposited on Fe(110) and successfully oxidized to AlO_x. The step-by-step oxidation of thin Al layers at room temperature leads to the formation of amorphous AlO_x on top of the Fe(110) surface. A subsequent annealing at 250 °C of the oxidized 7-Å thick Al layer results in the formation of a well-ordered $Al_2O_3(0001)$ layer on the Fe(110) surface.

Keywords: Auger spectroscopy; LEED; Surface oxide

1. Introduction

The heart of a magnetic random access memory (MRAM) device is a magnetic storage cell based on a submicron-sized magnetic tunnel junction (MTJ) which consists of two ferromagnetic electrodes separated by a thin insulating layer. MRAM devices are expected to have an important commercial impact due to the combination of its non-volatility with the properties of the existing dynamic random access memory (DRAM), such as high storage density, high read and write speed, low power consumption and long lifetime.

In the 1970's, Jullière[1] discovered the spin-polarized tunneling between two ferromagnetic electrodes (Fe and Co) separated by a thin semiconducting (Ge) layer and showed that tunneling magnetoresistance (TMR) depends on the relative orientation of the magnetic moments (magnetization directions) of the electrodes. However, only substantial improvements achieved in thin film growth and vacuum technology allowed in the 1990's the fabrication of junctions with reproducible characteristics and large TMR values [2,3].

The first theoretical explanation of the TMR effect was given by Jullière[1] within a simple model which assumes a spin-conserved tunneling process and in which the tunneling probability is proportional to the total electronic density of states in each electrode. In this model, the conductance is larger for parallel magnetized electrodes than for the antiparallel alignment [1]. However, the Jullière's model has several shortcomings for an adequate quantitative description of the spin-polarized tunneling process, such as neglecting the real electronic spin-polarized band structure of the electrode materials and of the insulator, hybridization at the interfaces as well as bulk and interface imperfections.

Recent *ab initio* calculations have shown that the magnetoelectronic properties of an MTJ strongly depend on the structural as well as electronic properties of the insulating layer and the specific termination at the insulator/ferromagnet (I/FM) interface [4–7].

In present, thin amorphous aluminium oxide (Al_2O_3) layers are typically used as insulating barriers in MTJs. For $Co/Al_2O_3/Co$ tunnel junctions, recent theories predict a positive spin polarization (SP) for the tunneling electrons because at small voltages the electrons with sp-hybridization tunnel more effectively than electrons with d-character [6]. However, changes in the sign of the SP of Co from plus to minus have been observed by changing Al_2O_3 to $SrTiO_3$ barriers [8].

In general, controlled epitaxial growth of ultrathin epitaxial Al_2O_3 layers on 3d ferromagnets with a well defined

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ferromagnet/insulator interface is of crucial importance for the understanding of the spin-dependent tunneling process in such systems.

Alumina, Al_2O_3, occurs in several structurally inequivalent crystallographic phases. The two most common and important phases are α-Al_2O_3 and γ-Al_2O_3. α-Al_2O_3 or sapphire, exhibits a trigonal structure known as corundum with Al^{3+} ions octahedrally coordinated by oxygen. The basic structural properties of γ-Al_2O_3 are not very clear yet. The present understanding is that γ-Al_2O_3 is a defective spinel with vacancies on cation sites and two types of Al coordinations: octahedral (AlO_6) and tetrahedral (AlO_4).

Recently, epitaxial α-Al_2O_3(0001) films have been prepared on different metal surfaces including Pt(111), Re(0001) and Ru(0001), for which the in-plane lattice mismatches with the oxygen sublattice of α-Al_2O_3(0001) are $> 0.79\%$, $> 0.43\%$ and $< 1.75\%$, respectively. However, these substrate surface orientations are also good candidates for the nucleation of γ-Al_2O_3(111), since both γ-Al_2O_3(111) and α-Al_2O_3(0001) exhibit a close-packed hexagonal oxygen sublattice with an O–O interatomic distance of 2.77 Å. The molecular-beam epitaxy growth of α-Al_2O_3(0001) on Pt(111) has not been described in the literature, although the inverted epitaxial system, Pt/α-Al_2O_3(0001), has been successfully grown by ion-beam sputtering [9]. Wu et al. [10–12] have grown epitaxial Al_2O_3(0001) films on Re(0001) and Ru(0001) by evaporation of Al in UHV followed by a post-annealing in O_2 as well as by evaporation of Al in O_2 at elevated substrate temperatures. Thin films of α-Al_2O_3 were also prepared on Ta(110) despite the fact that there is no simple epitaxial relationship between the two surfaces leading to a rotated Kurdjumov–Sachs orientational relationship [13]. In the α-Al_2O_3/Ta(110) system, a considerable in-plane expansion in the close-packed oxygen layer (3.05 Å compared to 2.77 Å was observed in the surface of bulk α-Al_2O_3(0001)), indicating the presence of significant strain.

A common procedure for the epitaxial Al_2O_3 film growth on metal surfaces is the ultra high vacuum deposition of Al in an oxygen atmosphere at elevated temperatures with or without subsequent annealing in oxygen [14,15]. This procedure yelds Al_2O_3 thin films which have structural and electronic properties close to those of bulk crystalline Al_2O_3. The main disadvantage of this growth procedure is the fact that the deposition of Al does not occur on the pure metal surface but on the partially oxidized or completely oxidized metal surface. This

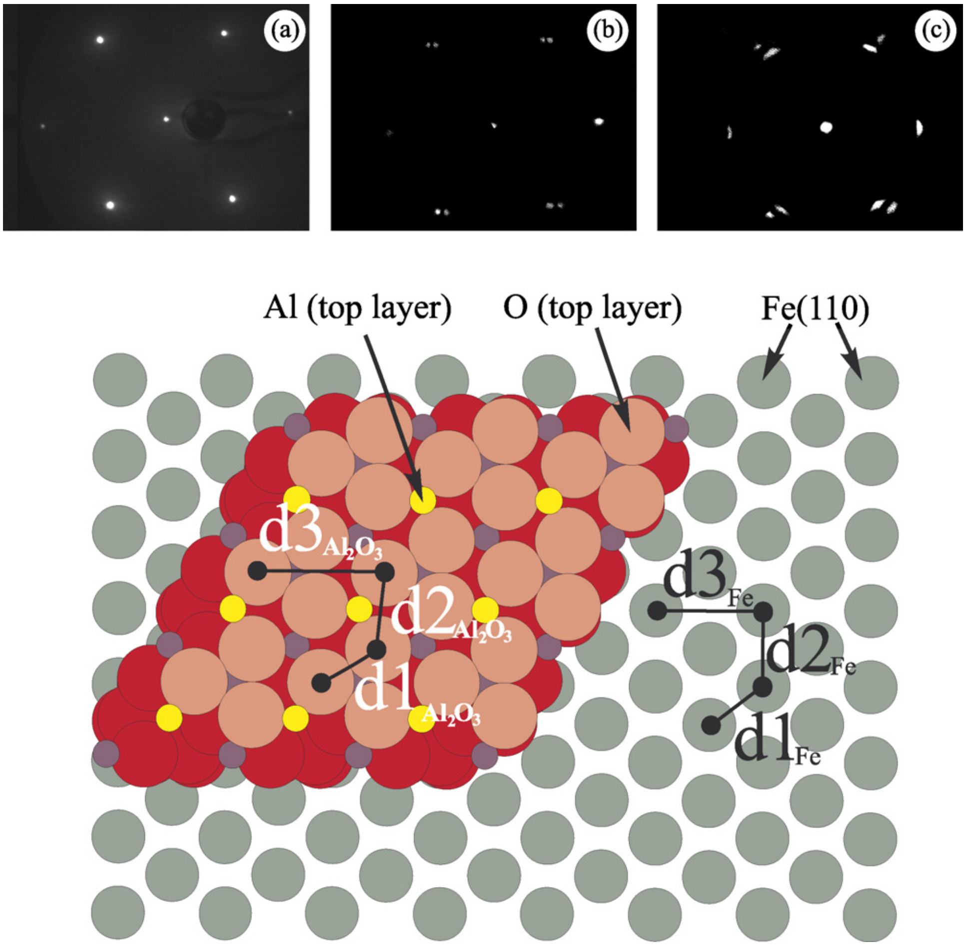

Fig. 1. Upper panel: LEED images of (a) 50-Å thick Fe(110) film on W(110) and 7-Å Al on Fe(110) before (b) as well as after oxidation (c) by 10^5 L O_2 exposure followed by annealing at 250 °C for 15 min. Lower panel: schematic presentation of a possible growth mode of 1 ML of Al_2O_3(0001) on top of the Fe(110) surface. The distances marked in the schematics are: $d1_{Al_2O_3} = 2.50$ Å, $d2_{Al_2O_3} = 2.88$ Å, $d3_{Al_2O_3} = 4.76$ Å and $d1_{Fe} = 2.48$ Å, $d2_{Fe} = 2.88$ Å, $d1_{Fe} = 4.05$ Å, $d2_{Fe} = 2.48$ Å, $d3_{Fe} = 4.05$ Å.