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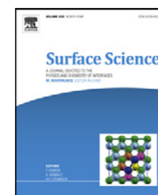

Surface Science Letters

Oxidation of Mg atomic monolayer onto silicon: A road toward $MgOx/Mg_2Si$ (11–1)/Si (100) heterostructure

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ABSTRACT

Surface interfaces of thin magnesium oxide films elaborated onto Si(100)–(2 × 1) substrates were characterized using scanning tunneling microscopy and spectroscopy, Auger electron spectroscopy, atomic force microscopy, and high-resolution transmission electron microscopy. We report that a flat and highly homogeneous magnesium oxide with well-defined interfaces could be grown at room temperature (RT) by repeating alternate adsorption of Mg atomic monolayer and O_2 on Si(100). RT oxidation process of the first Mg monolayer plays a crucial role as driving force allowing a partial decomposition of amorphous ultra-thin Mg_2Si at the Mg/Si interface to form more magnesium oxide in the surface. This process induces crystallization of the interfacial Mg_2Si thin film and then gives rise to an unexpected $MgOx/Mg_2Si(11–1)/Si(100)$ heterostructure. MgOx monolayer displays a band gap of about 6 eV and exhibits a weak RMS roughness on large areas.

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1. Introduction

For relevant applications in nanotechnology-related fields such as nanoelectronics, spintronics, molecular electronics, or sensoristics, e.g. [1–4], the ability to engineer ultrathin oxide layers mastering simultaneously chemical purity, homogeneity, and interface quality appears as a key issue [5–7]. More precisely, special attention must be paid to the structure of the interfacial layer between oxide and substrate, since interfacial defects could generate band gap energy states resulting in electron capture, namely, loss of polarization [8]. Amid the wide range of thin oxide films that have been extensively studied, magnesium oxide has attracted a growing interest over the past few years thanks to its high potentiality as a high-k dielectric material for electronic devices [6] or as an insulating barrier for magnetic tunnel junctions [9–11]. Indeed, many attempts have been made to reach high-quality MgO thin films onto silicon (100) and (111) surfaces.

However, despite the large panel of growth processes widely investigated (such as magnetron sputtering [5,8], physical layer deposition [12–14], molecular beam epitaxy [15,16], E-beam [17–19] etc.), some weighty drawbacks inherent to the lack of control of the interface formation still remain. In particular, recent works report the formation of an amorphous silicon oxide layer at the MgO/Si interface due to subcutaneous substrate oxidation [5], as much as the existence of an uncontrolled intermixing layer in the case of MgO deposited on hydrogenated Si surfaces [12,16].

In order to overcome such technological locks, some recent studies explore an innovating growth method, called ALDO for atomic layer deposition and oxidation, in which the oxidation is performed at RT on a metallic monolayer (ML) previously deposited on silver or silicon substrate [20–25]. In particular, ALDO process [26–28] was successfully used to elaborate aluminum oxide onto silicon surface, resulting in high homogeneous oxide layer with abrupt interfaces and preventing the oxidation of the silicon substrate [26].

Based on alternate adsorption of Mg atomic monolayer and O_2 exposure at RT, we report here the formation of ultra-pure and homogeneous ultrathin magnesium oxide layer onto Si(100). Supported by ex-situ TEM characterization, we highlight that RT oxidation process of the first Mg ML induces crystallization of a thin Mg_2Si interfacial layer, resulting in a Si(100)/Mg_2Si (11 − 1)/amorphous MgOx layers stack. Regarding to the high-quality of this achieved interfacial layer (i.e. low concentration of electrically active defects, abrupt interfaces and particularly no silicon oxide formation), such architecture could be of great interest in sense of electronic transport properties.

2. Experimental process

In-situ experiments were performed in an ultra-high vacuum (UHV) chamber equipped with a commercial Omicron variable temperature-scanning tunneling microscopy (VT-STM), an Omicron Spectra low-energy electron diffraction (LEED), and a Riber CMA Auger spectrometer dedicated to surface analyses. Additional ex-situ analyses were

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performed using an environmental JEOL atomic force microscopy (AFM) operating in a noncontact mode for roughness measurements, and a high-resolution transmission electron microscopy (HR-TEM) for interfaces characterization.

Prior to elaboration process, the Si(100) samples were chemically cleaned, introduced into the UHV chamber, and outgassed at 700 °C overnight. Afterward, and in order to remove the surface oxide, the samples were briefly flashed at 1100 °C before the temperature was rapidly brought to 900 °C; the samples were then slowly cooled (around 15 °C/min) down to 650 °C.

This procedure was reiterated until the Si(100) surface was reconstructed in the (2×1) pattern: a sharp (2×1) LEED pattern and STM images of the (2×1)-reconstructed surface were thus obtained.

Ultra-thin magnesium oxide layers were then elaborated using the following procedure: (*i*) RT deposition of one magnesium ML from a calibrated effusion cell at a background pressure of 4×10^{-10} Torr, (*ii*) RT oxidation of the Mg ML by exposure to a constant pressure of molecular oxygen (30 Langmuir) in the UHV chamber. AES growth curves obtained from our preliminary investigation of the Mg onto Si(100) system [29] were used as a reference for the calibration of the deposition rate. In the following discussion, note that one monolayer corresponds to the atomic density of the Si(100) surface layer, i.e. 7.88×10^{14} atoms/cm^2.

For TEM analyses, we obtained a flattened surface thanks to the modified Shiraki treatment and the repeated surface reconstruction flashes preceding the elaboration stage.

3. Results and discussion

Fig. 1(a) displays AES spectra recorded at constant emission current for clean silicon, after deposition of one magnesium monolayer onto the substrate and as a consequence of oxidation process followed by equilibrium annealing (450 °C). The first spectrum shows a Si_(92 eV) substrate peak completely clean after the sample preparation. On the second spectrum, a Mg_(45 eV) peak appears and contributes to the attenuation of the silicon peak by about 54% of its initial value. One can note that the peak-to-peak Auger intensities ratio between substrate and adsorbate is close to 1. This value was previously identified [29] as corresponding to the deposition of one magnesium ML onto the substrate. The third spectrum recorded after 30 L of O_2 exposure and UHV annealing shows the appearance of an extra O_(510 eV) peak. Regarding to the attenuation of both Mg and Si intensities after oxidation, one can suppose that the oxygen atoms are adsorbed on top of the magnesium ML. From the 90% total attenuation of the Si peak measured after Mg deposition and oxidation, we extract using attenuation equations [30,31] a thickness of 8 Å for the deposited MgOx ML. Fig. 1(b) shows more clearly the evolution of the magnesium peak due to the oxidation process. After deposition of the metallic ML, magnesium signature appears as a Mg_(45 eV) peak which corresponds to atoms in a metallic state [32]. As shown in the second spectrum, oxidation process changes the chemical environment of the metallic ML and leads to a noticeable magnesium peak shift of 10 eV toward lower energy, resulting in the appearance of an oxidized Mg_(35 eV) peak. On the other hand, no significant changes in terms of shape or energetic position can be seen for the Si_(92 eV) peak before and after oxidation. From this observation, we can assume no oxygen diffusion through the magnesium layer during oxidation process followed by 450 °C equilibrium annealing. This observation is, however, strongly consistent with our previous report about the formation of an interfacial magnesium silicide layer whose self-limited growth at Mg saturation of the silicon surface seems to act as diffusion barrier for the oxygen atoms [29] and prevents oxidation of the Si substrate.

The RT oxidation process and its impact on the resulting oxide surface morphology were investigated using STM.

Fig. 2(a) displays a filled states STM image of the surface after deposition of one magnesium ML. The silicon substrate appears completely covered by a non-continuous magnesium film that organizes itself as homogeneous 2D flakes with a lateral size in the 10–20 nm range. Fig. 2(b) presents a filled states STM image recorded after exposing the magnesium monolayer to 1.5 Langmuir of O_2. This weak exposure was firstly controlled by AES, resulting in the coexistence of both metallic Mg_(45 eV) and oxidized Mg_(35 eV) signatures, so that the early stages of oxidation process could be highlighted. One can still observe the global shape of the magnesium flakes whose edges appear now decorated with a high quantity of subnanometer dots and reasonably attributed to oxidation effect. From this observation, we can assume that oxygen adsorption seems driven by a surface reactivity process taking place on top of each Mg flake, from the periphery to the center. Effectively, we note at very low exposure (1.5 L of O_2) that Mg flakes center stay flat and homogeneous (black uniform color) while the topography of periphery changes. Fig. 2(c) exhibits a filled states STM image recorded after 30 L of oxidation followed by 15 min UHV annealing at 450 °C. In good agreement with the loss of the metallic Mg_(45 eV) Auger signature at this exposure (spectrum not shown), a stronger oxidation changes the surface morphology of the layer by increasing significantly the density of subnanometer dots reasonably similar to the ones observed after oxidation of a Mg monolayer on Ag(111) [22]. This complete oxidation followed by equilibrium annealing results in the formation of a dense and homogeneous ultra-thin oxide layer with low surface roughness, according to the 5 Å average height obtained from the corresponding height profile.

Additional ex-situ AFM experiments (not shown) were performed onto one annealed MgOx ML, resulting in a root-mean-square (RMS) roughness measurement as low as 6.8 Å (300×300 nm^2 scan area),

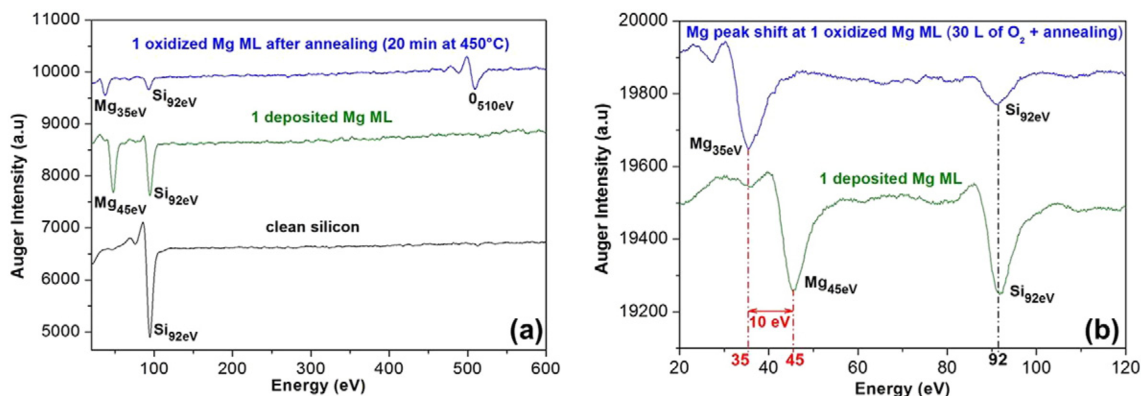

Fig. 1. (a) AES spectra recorded at constant emission current for clean silicon, after deposition of one magnesium monolayer onto the substrate and as a consequence of oxidation process followed by equilibrium annealing at 450 °C. (b) Focus on magnesium and silicon Auger signatures observed in (a) after 1 Mg ML deposited and as a consequence of oxidation followed by annealing.

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