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Journal Article

N.B.: When citing this work, cite the original article.

Original Publication:
http://dx.doi.org/10.1116/1.4975595
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Postprint available at: Linköping University Electronic Press
http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-136638
Wet-cleaning of MgO(001): modification of surface chemistry and effects on thin film growth investigated by X-ray photoelectron spectroscopy and time-of-flight secondary ion mass spectroscopy

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The effect of the wet-cleaning process using solvents and detergent on the surface chemistry of MgO(001) substrate for film deposition was investigated. Six different wet-cleaning processes using solvent and detergent were compared. The effect on film growth was studied by the example system ScN. The surface chemistry of the cleaned surface was studied by X-ray photoelectron spectroscopy and the film/substrate interface after film growth was investigated by time-of-flight secondary ion mass spectroscopy. The composition of the surface is dependent on the wet-cleaning process. Sonication in a detergent before the solvents yields a pure oxide surface compared to hydroxide / carbonate contaminated surface for all the other processes. An annealing step is efficient for the removal of carbon contamination as well as most of the hydroxide or carbonates. The study of the film/substrate interface revealed that the wet-cleaning process significantly affects the final interface and film quality. The substrate cleaned with detergent followed by solvent cleaning exhibited the cleanest surface of the substrate before annealing, after annealing and also the sharpest film/substrate interface.
I. INTRODUCTION

Preparation and cleaning of the substrate is the first and most important step before thin film deposition. Substrate preparation starts from cutting and polishing of single crystal to very complex structures achieved by lithography techniques with purposes such as the removal of contaminants and native oxide layers. Many different processes are used to prepare and clean the substrate. In most cases, combinations of different techniques are used.

This paper focuses on the cleaning processes of the substrate. A non-exhaustive list of cleaning techniques is listed in Table 1 and divided in two categories: wet-cleaning and dry cleaning techniques. The purpose of cleaning the substrate is essentially to reduce and ideally remove all surface contaminants before deposition. Sonication in commonly used solvents such as acetone, ethanol and isopropanol is the most reported approach in the literature. The strongest wet-cleaning process is the use of acid in order to remove a surface layer. Another process, more common in electronic-component manufacturing, is the process using any soap or detergent efficient against dust, grease and lubricant. Among dry cleaning processes, annealing at high temperature (> 500°C) is the most used among the thin film community. Annealing removes the main organic contaminants and may change the surface by atomic reconstruction of the surface if the temperature used is high enough. The other techniques are soft etching (UV-ozone cleaner) and plasma ion etching (ex-situ or in-situ) used to reduce or remove the surface layers (contaminants or native layers).
The present paper focuses on magnesium oxide (MgO) substrates, which are widely used in thin film technology for applications such as high temperature superconductors, magnetic, piezoelectric, optoelectronic and thermoelectric materials. MgO is also a common substrate used for the development and research on new material systems. MgO single crystal is suitable for cube-on-cube deposition, tetragonal materials or hexagonal materials on (111)-oriented substrates. Control of the film/MgO substrate interface is essential for the different applications mentioned and growth of new materials. Therefore, good control of the cleaning step before deposition is necessary.

The production and the control of the quality of this kind of substrate remain challenging. Schroeder et al. emphasized the difficulties to obtain a “single crystal” of MgO. The crystallographic quality of the substrates between suppliers varies from a mono-domain wafer to wafer containing multiple domains (for 70% of the MgO substrates studied). The other problematic aspect of this material is that MgO substrate is sensitive to moisture due to its hygroscopic character. This results to a surface reaction of the oxide forming hydroxides and/or carbonates. MgO is normally packed in an inert gas atmosphere and individually sealed in a bag to reduce the exposure to moisture while storing. The substrates are usually exposed to the ambient only prior to the deposition with an initial cleaning step, but in most cases the surface of commercially available magnesium oxide substrate is contaminated by water and/or hydro-carbons.

The cleaning of MgO substrates was extensively studied with the development of new high temperature superconductors (1995-2005). Different techniques have been used to remove contaminations, such as sonication in common solvents, in-situ annealing, plasma etching, and chemical etching (Table 1). Most of these studies showed the effectiveness of different processes on the removal and control of the surface contaminants, focusing their investigations on the removal or reduction of the carbon at the surface. Some papers presented X-Ray photoelectron spectroscopy (XPS) analysis results of C1s and O1s core-level peaks, but unfortunately not from the Mg2s which
may confirm the presence of hydroxide or carbonates of magnesium left at the surface $^{12,22}$. For example, Du et al. reported the effectiveness of the plasma cleaning before the deposition of YBCO where the quality of the film was greatly improved using the in-situ plasma etching $^{22}$. Tolstova et al. claimed improvement of thin film deposition and epitaxy using pre-cleaning or backspattering using RF substrate bias prior to deposition $^{23}$. The carbon contaminants were eliminated and the chemical environment of the oxygen modified; nevertheless, the magnesium bonding was not investigated by XPS.

The most reported wet-cleaning procedures is sonication in acetone followed by isopropanol and in some cases an in-situ annealing at high temperature in the deposition chamber. The difficulties and/or the lack of evidence concerning removal of hydroxides and carbonates from the surface of MgO substrate remains a problematic issue for the use of MgO substrates for thin film deposition.

In this paper, X-ray photoelectron spectroscopy (XPS) is used to address the surface chemistry of MgO(001) substrate subjected to various pretreatments. The bare substrate presented a full contaminated surface with mainly hydroxides and carbonates. The effect of the different wet-cleaning processes using a combination of common solvents, heptane and detergent on the surface composition is discussed. The same cleaning processes associated with an in-situ annealing at high temperature is also investigated. Finally, the last treatment procedure is used in real conditions with an example of the deposition of a ScN thin film with the effect on the quality of ScN crystallinity and film/substrate interface studied by time-of-flight secondary ions mass spectroscopy (ToF-SIMS).
II. EXPERIMENTAL SECTION

A. Preparation of the samples

MgO(001) substrates with a size of 10×10 mm² and (Alineason Materials Technology) were individually sealed and stored in a desiccator before any experiment. The reference sample, labeled REF, was only blown with nitrogen gas. Different sonications in solvents were used for the wet-cleaning process and dry under a nitrogen flow at the end: acetone (10 min) + isopropanol (10 min) (labeled AI); Acetone (10 min) + Ethanol (10 min) (labeled AE); Heptane (10 min) + Acetone (10 min) + Ethanol (10 min) (labeled HAE). The soap/detergent used in this study was the Hellmanex®II diluted in the recommended proportion (2 vol-%). A Hellmanex®II alkaline concentrated detergent (K₃PO₄ 15-30%, anionic surfactants < 15% and non-anionic surfactant < 5%) is a commercial cleaning solution used to wash cuvettes and widely used for cleaning glass substrates. A detergent with similar composition or sold for the same purpose should give similar effects as the ones observed in this study. The substrates were washed with a sonication in detergent for 3 min and subsequently rinsed twice in de-ionized water during 5 min (labeled D). Finally, a combination of detergent and solvent was used with the detergent “method” followed by acetone for 10 min, then ethanol for 10 min (labeled DAE). The same wet-cleaning procedures were used before an annealing step at 700°C/2h in a deposition chamber with a base pressure of 10⁻⁸ torr. To assess effects on film growth a 150 nm thick film of ScN were deposited on MgO(001) substrate by DC magnetron sputtering operating at 120 W under 0.27 Pa (2 mtorr) total pressure with 25 %N₂ and with a substrate temperature of 950°C during 75 min. The substrates were cleaned prior to deposition by the different wet-cleaning procedures described above followed by in-situ annealing at 700°C/2h under vacuum (10⁻⁸ Torr). For brevity and readability, abbreviations are used for the 18 samples and are listed in Table 2. The cleaning of the sample was performed just prior the different characterizations with a minimum time (< 3 min) between the
cleaning and insertion in the XPS and ToF-SIMS chamber or the contact angle measurement (the samples were kept sealed in a box for transportation). To keep that restriction of time between cleaning and analyses and avoiding any more contamination on the surface, new substrates were taken and the cleaning were repeated for the three parts of the study (wet-cleaning / annealing / deposition).

**B. Characterization**

1. **Contact angle measurement**

   The contact angle measurement was measured using CAM 200 optical contact angle meter (KSV Instruments Ltd., Helsinki, Finland). A droplet of approximately 5-7 μL was deposited on the surface using a syringe. The contact angles were computed based on the images of droplet on the MgO surface. For each sample a minimum of four different readings were recorded. A typical error in readings was ~2°.

2. **X-ray photoelectron spectroscopy (XPS)**

   XPS spectra were obtained using an Axis Ultra DLD instrument from Kratos Analytical (UK) with the base pressure during spectra acquisition of 1.1×10⁻⁹ Torr (1.5×10⁻⁷ Pa), with a monochromatic Al Kα radiation (hv = 1486.6 eV). The anode power was set to 225 W. All spectra were collected from the area of 0.3×0.7 mm² and at normal emission angle. The analyzer pass energy was set to 20 eV which results in the full width at half maximum of 0.55 eV for the Ag 3d₅/₂ peak.

   XPS data were treated using KolXPD fitting software²⁴. The C1s peak located at 284.8 eV was used as the charge reference. Peak fitting can be done in different ways and the results will, therefore, depend on the chosen constraints applied in the deconvolution process. Peak fitting was performed for all photoelectron peaks with Voigt function after a Shirley background subtraction. The data treatment
was done from the simplest to the more complex spectra by fixing constraints as described in the appendix.


Elemental distributions in the films were measured by time-of-flight Secondary Ion Mass Spectrometry (ToF-SIMS) using a ToF.SIMS V instrument (ION-TOF GmbH, Germany). Dual beam depth profiling, by alternately applying an analysis beam and a sputter beam (non-interlace), was done in positive and negative secondary ion mode. Here, selected positive respective negative secondary ion species were monitored as a function of sputter time. Low-energy electron flooding was applied for charge-compensation during profiling.

For the positive mode a quasi-continuous 2.0 keV O$_2^+$ beam with a current of 505 nA and scanned over 300 x 300 µm$^2$, was used as the sputter beam. In the negative mode, a quasi-continuous 2.0 keV Cs$^+$ beam, having a current of 137 nA and scanned over 330 x 330 µm$^2$, was used as the sputter beam. In both positive and negative ion mode a pulsed 30 keV Bi$^+$ beam, cycle time 50 µs, was used as the analysis beam, with a target current of 3.0 pA and an analysis field of view of 70 x 70 µm$^2$ at the centre of the sputter craters. Ion mass spectra were acquired, with an extraction voltage of (-/+2000 V between the sputter sequences, in the so-called spectroscopy mode (bunched 6.5 ns Bi$^+$ ion beam pulse width, mass resolution m/Δm ~ 6000-8000, beam spot ~5 µm). SurfaceLab 6 software (v. 6.5, ION-TOF GmbH) was used for spectra recording and data processing.

4. **X-ray diffraction (XRD)**

For XRD analysis, two diffractometers were used: a PANalytical X’Pert PRO in Bragg Brentano configuration (with continuous rotation of the sample during the measurement) equipped with
a Cu K$_\alpha$ radiation source and a four-circle texture instrument (PANalytical X’Pert MRD) equipped with a parallel beam Cu K$_{\alpha 1}$ radiation in $\theta$–$2\theta$, $\omega$, and $\phi$ scan modes.
III. Results and discussion

The paper is divided into three sections corresponding to the control of the surface from the moment the MgO substrate is out of the box to the final film/substrate interface. The analyses of the surface are performed after: (a) wet-cleaning only, (b) after subsequent in-situ annealing, and (c) after ScN film growth (analyses of the ScN/MgO interface).

A. Wet-cleaning treatment on MgO(001) substrate

This section focus on wet-cleaning only. Some deposition techniques such as solution deposition by spin coating and dip coating do not use any vacuum or high temperature processes before the film deposition performed. The control of the surface contamination/properties is important to control thickness homogeneity, compositions and properties of the final film after annealing in the later steps. The composition of the surface before insertion in the vacuum chamber will affect the surface after annealing and/or the interface thin film/MgO.

Five different wet-cleaning processes were compared with the reference sample using contact angle measurements of a drop of water on the MgO surface and XPS analysis. The contaminated surface of the reference substrate became cleaner after wet-cleaning. The choice of a proper wet-cleaning solution significantly affects the wettability and the composition of the surface up to the recovering of the pure oxide at the surface.

Figure 1 shows photographs of the side view of the water droplet deposited on the surface of MgO(001) substrate for the five different wet-cleaning treatments used in the present paper. The as-received sample (labelled REF) had a hydrophobic surface with a contact angle of 80°. The cleaning with the organic solvent only (fig1-b, c and d) did not improve the wettability of the surface, yielding an even higher contact angle of 96° for the AI sample. The use of a detergent clearly decreased the
contact angle to a minimum of 15° for the D sample and 32° for the DAE sample. The surface of MgO cleaned using a detergent was found to be hydrophilic. This change of wettability of the surface showed that contaminants (particles, oil, wax, hydroxides, carbonates …) present on the as-received substrate (REF) was removed after a wet-cleaning process using a detergent. Wettability of the substrate is an important aspect for the chemical solution coating using spin/dip-coating or spraying of a film with homogeneous thickness. A hydrophilic surface is preferred for a good spread of the resin on the substrate, therefore a detergent step may be necessary to improve the deposition by solution on MgO(001).

In order to identify the surface chemistry of the substrate surface, XPS analysis was performed after each treatment. Figure 2 shows the XPS analysis of the Mg2s peak of the surface of the six samples and a summary of the composition of the surface evaluated after fitting of the Mg2s peak. Figure 3 shows the XPS analysis of the O1s peak and a summary of the composition estimated from the O1s peak subtracting the contaminant component (adsorb gases) of the same samples.

The fit of the Mg2s peak from the REF sample (Figure 2-a), revealed four components identified from the lowest to the highest binding energy as: MgO (87.7 eV), Mg(OH)2 (88.4 eV), MgCO3 (89.2 eV), and a component at higher binding energy (90.2 eV) due to a group or element bonded to the magnesium with a higher electronegativity than O, OH or CO3. This group may be COOH (CHMgO2). No trace of fluorine, chlorine, aluminum, or calcium was detected by XPS. The same components were found in the O1s peak (figure 3a): an oxide component at 529.4eV; a common hydroxide/carbonate component; a C=O component at 531.5 eV (related to the high bonding energy component on Mg2s); a contamination component from adsorbed gases (subtracted in the calculation); and a component at higher binding energy which may correspond to H2O at 533.9eV. The
oxide/(hydroxide + carbonate) ratios extracted from the two core-level peaks are similar (figure 2-g and 3-g).

The surface of the AI sample had less contaminants than the REF sample (figure 2-b). Cleaning by acetone following by isopropanol removed the contaminants responsible for the peak for Mg2s at high binding energy but the relative ratio of oxide/contaminants remains approximately the same as before the cleaning (figure 2-g). The same observation was made on the O1s peak with the absence of the component at 529.2 eV and another component at 533.0 eV which may issue from the left over of the solvent and corresponding to C-O-H or C-O-C.

The Mg2s peak and the O1s peak of the surface of the sample AE had only two components (without the contamination peak on the O1s peak): approximately 60% of MgO and 40% of Mg(OH)$_2$ (figure 2-c and g).

Adding an extra sonication step in heptane prior to the ones in acetone followed by ethanol improved the cleaning process (figure 2-d/g and 3-b/g), with a final surface composed with approximately 96 % of oxide. The 4 % of magnesium hydroxide remaining at the surface of the sample HAE could correspond to few monolayers of Mg(OH)$_2$.

The detergent process (sample D) eliminated the main contaminants on the as-received substrate (identified by the peak at high binding energy on Mg2s), while the carbonates and hydroxides were partially removed (figure 2-e and g). The final surface was composed with 45 % of magnesium oxide compared to 19 % before. The O1s core-level peak is composed with several components similar to the one observed on the REF sample but non observable on the Mg2s peak. The different components of the O1s peak (figure 3-e and g) at 531.5 eV (C=O), 533.0 eV (C-O-C or C-O-H) and 533.9 eV (H$_2$O) are from the left over from the detergent and/or water used in the process without bonding to the magnesium.
In the last process, the combination of the detergent and the common solvents (acetone / ethanol) gave the best results with a surface recovered with only magnesium oxide (figure 2-f and g). The detergent interacts with the bonding of the hydroxide and the carbonates on the surface of the substrate, which then are eliminated by the two steps of sonication in acetone and ethanol. The same observation was made on the O1s (figure 3-f and g).

Figure 4 shows the XPS analysis of the C1s peak of the six different samples after wet-cleaning treatment. The REF sample had a broad C1s peak which includes different components such as the C–C (285 eV), C–H (285 eV), C–O (286 eV), C=O (287-288 eV) issued from the different contaminants at the surface identified on the Mg2s peak plus the extra carbon from adsorbed gas and particles. The peak at low binding energy may correspond to dehydrogenated carbon species bonded to a metal ion. The different treatments affected the carbon peak C1s by eliminating the component at higher binding energy (C=O) which is consistent with the observation on the Mg1s and O1s peak. The narrowest peaks of carbon were observed for the sample AE and DAE with a main component C–C issue, most probably from the leftover after the evaporation of the ethanol. The samples AI had a high amount of carbon at the surface with a wider peak of C1s explained by the presence of carbonates and hydroxides higher on its surface. The C1s spectra of the surface of the sample D confirmed the analysis on the Mg2s peak with the presence of carbonates and hydroxide. The component C–O at 286 eV was present and the amount of carbon was lower than the sample D because no solvent was used at the last step of the cleaning. The sample HAE had the lowest amount of carbon at the surface even with the use of ethanol at the last step of the cleaning in contrast with the sample D where the C–C component increased after the solvent step.

To conclude this section, the as-received substrate had a fully contaminated surface with hydroxides, carbonates etc. The different wet-cleaning processes changed the surface chemistry by removing the different contaminants with a variable effectiveness. The DAE samples had a pure oxide
layer covering the substrate while the other samples had hydroxides and carbonates with different proportions after the wet-cleaning. The amount of carbon detected at the surface of the sample was not negligible, and was brought by the leftover of solvent at the last step of the process. The wettability was improved with a minimum contact angle of 15° for the D sample and 32° for the DAE sample which can be an important parameter for a deposition by dip or spin-coating.

**B. In-situ annealing treatment on MgO(001) substrate after the wet-cleaning treatment**

This section focus on the annealing effect on surface composition. The wet-cleaning by solvents followed by in-situ annealing is the most common reported procedure used when depositing on MgO. Here, the surface of the six different samples (REF + the five wet-cleanings (see section 1)) were analyzed by XPS. For the composition, the annealing change the surface chemistry at the surface with a final surface composed of between 80 % and 100 % of magnesium oxide regarding the wet-cleaning chosen.

The same cleaning processes were used to clean the substrate before an annealing step under 10⁻⁸ mTorr at 700 °C during 2 h. Figure 5 shows photographs of the substrate of magnesium oxide after annealing compared with the as-received substrate. A color gradient is observable between samples from transparent to grey/brown. The sample AI and AE presented the largest change of color with a grey/brown tint. The sample without any wet-cleaning treatment had a light grey tint. For the 3 others (HAE-A, D-A, and DAE-A), no significant change of the color was observable. This change of optical properties is not clearly understood. However the change can be correlated with the amount of carbon at the surface before annealing (figure 4). The samples exhibiting a change of color are the ones having
the highest amount of carbon left on the surface before the annealing step. In addition, other studies show that defects in MgO may affect the optical properties.$^{26,27}$

Figure 6 shows the XPS analysis of the Mg2s peak of the surface of the six samples after annealing and a summary of the composition of the surface evaluated after fitting of the Mg2s peak. Again no trace of fluorine, chlorine, aluminum or calcium was detected by XPS. Each surface had the MgO as the main component of the Mg2s peak with different ratios regarding the cleaning process. The sample DAE-A had a pure oxide surface as it was before annealing (figure 6-f and 6-g). The HAE-A and AE-A sample had a surface composed with 98% and 91% of oxide, with few percent of magnesium hydroxide (figure 6-c, d and g). The sample REF-A and D-A showed similar results with a surface composed at approximately 95% of oxide (figure 6-a, e and g). Finally, the XPS analysis of the surface of the AI sample revealed 3 components: MgO (80%), Mg(OH)$_2$ (14%) and MgCO$_3$ (6%) (Figure 6-b and g).

Figure 7 shows the XPS analysis of the O1s peak and a summary of the composition estimated from the O1s peak subtracting the contaminant component (adsorbed gases) of the MgO surface after the six different wet-cleaning processes followed by annealing at 700 °C for 2h. The six different samples have a contamination component (20 % of the full area) explained by the short time between the deposition chamber and XPS chamber where adsorbed gases may attach to the surface. The analyses of the O1s core level peak are in accordance with the observation made from the investigation of the Mg2s peak. The percentages of the different components after subtracting the contamination component are similar within the error as the one calculated from the Mg2s peak. The sample AI presented an extra component (3%) at high binding energy (533.1 eV) and may correspond to a bond related C-O-C or C-O-H, which also was present prior to the annealing and seemed to resist the annealing step.
Figure 8 shows the XPS analysis of the C1s peak of the six different samples after the annealing treatment. All the six samples had a similar C1s spectra with approximately the same amount of carbon at the surface. Annealing of the sample drastically decreased the amount of carbon present at the surface (figure 4 and 7). The small amount of carbon detected at the surface may be due to species adsorbed at the surface during the short transfer time between the annealing chamber and insertion in XPS chamber.

In-situ annealing of samples at 700°C/2h under vacuum decreased the contaminants, but a few percent of magnesium hydroxide remained. With an XPS analysis depth of 50 - 100 Å and a Mg-OH bonding of 3 - 4 Å, the detection of a monolayer of hydroxide well bonded to MgO at the surface would correspond to 3 – 8 % of the total signal. The decomposition of hydroxides and carbonates occurs both at a temperature higher than 350°C, but it has been demonstrated that the decomposition can only be fully achieved using temperatures higher than 900°C and 1000°C for magnesium carbonates and hydroxides, respectively. Annealing the samples at these temperatures may initiate a reconstruction of the surface, and a possibly segregation of the bulk impurities (mainly calcium) to the surface. In an application focusing on reducing the carbon contaminants, a procedure using sonication in acetone flowed by isopropanol would be considered a good process. However, the samples AI-A showed the worst results in terms of removing hydroxides and carbonates contaminants, which are left and visible on the Mg2s peak with only 80% of Mg oxide at the surface.

The annealing process affected the surface compositions of all samples with a significant reduction of contaminants. The presence of carbon, which was brought by the leftover from solvents was eliminated after annealing. The AI-A sample was the sample with the highest amount of contaminants (hydroxides and carbonates) with only 80 % of Mg oxide. The other samples had a
surface composed of oxide and hydroxides with a minimum of 90 % of MgO up to a 100 % of MgO for the surface of the DAE sample.

C. Deposition of ScN thin film on in situ-annealed MgO substrates

In the third and final section of this paper, we focus on ScN film deposition on the different substrates (the REF-A, AI-A, AE-A, HAE-A, and the DAE-A sample). The film/substrate interface was, after deposition, analyzed by ToF-SIMS. A clear difference between the samples was observed showing that choice of wet-cleaning seemingly influence the quality of the oriented thin film and resulted in diffusion between the substrate and the film. The DAE-F and AE-F samples presented the smallest diffusion range/width.

Figure 9 shows the diffraction patterns of the five samples of ScN/MgO(001). Note that all samples were deposited simultaneously, there is only a difference in cleaning procedure. All the thin films exhibited an epitaxial (001) ScN orientation \(^{30,31}\). The thin film AI-F and HAE-F present a secondary orientation of (111) ScN and a secondary phase (indicated by *), which can be identified either as the 222 of Sc\(_2\)O\(_3\) (cubic / \(a = 9.84 \text{ Å}\)) or 222 Mg\(_3\)N\(_2\) (cubic / \(a = 9.97\text{ Å}\)) at \(2\theta = 31.3^\circ\).

Table 3 presents the cell parameter along the growth direction and the FWHM of the \(\omega\)-scan and the \(\phi\)-scan corresponding the (001) ScN oriented grains for the five different samples. The crystallographic quality of the substrate (as quantified by FWHM) is also reported in the Table 3. The orientation quality of the grains, estimated by the values of FWHM of the \(\omega\)-scan and \(\phi\)-scan, decreases for the sample AI-F. The AI-F sample exhibited also a lower cell parameter along the growth direction compare to the four others samples. The small difference of the cell parameter of ScN may be due to a relaxed-lattice parameter with a film less ordered than the others samples which have a higher epitaxial
quality. A slight decrease of the quality of the MgO substrate, observed by an increase of the FWHM of the \( \omega \)-scan and \( \varphi \)-scan did not affect the quality of the 200 nm thick film of HAE sample.

For the ToF-SIMS analysis of the ScN films deposited on the differently cleaned MgO substrates, some characteristic masses were selected for the depth profiles. Some of the detected elements/masses in the thin film were intentionally not plotted for the clarity of the figures. The film contains for example a significant amount of oxygen (from residual water vapor in the chamber). In addition, there is a small amount of fluorine and chlorine in the films. Fluorine in an amount below 1 % has previously been detected in Sc containing films \(^{31,32,33}\) and is known to come from the sputter target. No sodium or potassium were detected at the interface (below detection limit). These are often seen as contaminant on surfaces by ToF-SIMS in the positive mode due to the high sensitivity for the alkali metals \(^{34}\). Different wet-cleaning procedures in addition to heating may change the amount of alkali metals (in the form of various salts) present at the surface.

Figure 10 shows the ToF-SIMS depth profile for the five different samples taken in positive and negative mode. Characteristic masses were chosen to identify the substrate (\(^{26}\text{Mg}^+\) and \(^{26}\text{MgO}^2\))_, the film (\(\text{Sc}^+, \text{ScN}^+, \text{Sc}^-\) and \(\text{ScN}^-\)) and the interface (\(\text{ScMg}^+, \text{Al}^+, \text{MgN}^-\) and \(\text{Al}^-\)). All profiles were normalized relative to the substrate signal of \(^{26}\text{Mg}^+\) or \(^{26}\text{MgO}^2\) in the positive and negative mode, respectively. The aluminum is located at the top surface of the MgO substrate and comes from the polishing of the substrates with alumina particles. In the positive mode, the high signal level observed for most ions in the substrate are not due to a diffusion but it is the background level in MgO.

In addition to the above masses, we also saw a peak due to \(^{40}\text{Ca}\). Calcium is known to be present as a trace element in MgO substrate \(^{15,29}\). The sensitivity of ToF-SIMS to \(\text{Ca}^+\) and \(\text{Al}^+\) is high compared to the other masses even when present in small amount. These elements were not detected (below detection limit) by XPS analysis of the surface of the substrate (section 1 and 2). The deposition
at 950°C may play a role on the diffusion of calcium into the film with a similar behavior as the magnesium diffusion into the film. It should be noted that even though the $^{40}$Ca mass peak ($m = 39.9626$) is well separated to the mass peak of $^{24}$Mg$^{16}$O ($m = 39.9799$), charging of the insulator MgO during depth profiling induces a slight mass shift in addition to a broadening of the mass peaks when entering the substrate. Although, a mass shift correction was applied in the subsequent data analysis, there could be cross contamination from MgO in the selected Ca mass range and vice versa. Therefore, one should be careful with comparing the levels of MgO/Ca in the substrate for the different samples. We nonetheless see similar behavior for the other Ca isotopes with regard to diffusion.

The REF sample exhibited a sharp interface between film and substrate with abrupt change in intensity for the Sc$^+$, ScN$^+$, Sc$^-$, ScN$^-$ of the film into the substrate. Nevertheless, a diffusion of magnesium and calcium was detected into the film. The AI-F sample exhibited a diffuse interface with a high diffusion of magnesium (see $^{26}$Mg$^+$, ScMg$^+$ and MgN$^-$) and calcium into the film and of scandium in the substrate (see Sc$^+$ and Sc$^-$). We note that the ScN has a more abrupt decrease in intensity. We see the the same feature for the N$^+$ signal (not shown here). The sample AE exhibited a similar case as the REF-F sample with a sharp film/substrate interface and a low amount of calcium diffusion into the film (Ca$^+$). The sample HAE-F had a diffuse interface with a diffusion of magnesium (see $^{26}$Mg$^+$, ScMg$^+$, MgN$^-$), calcium (Ca$^+$) diffusing into the film and scandium into the substrate (see Sc$^+$, Sc$^-$). Again, we note that the ScN$^-$ has a more abrupt decrease in intensity. The relative amount of magnesium diffusing in the film is lower than for the AI-F sample, but a higher diffusion of Sc in the substrate is observed with the presence of a plateau features. The sample DAE-F exhibited the sharpest interface indicated by Al, MgN and ScMg, with a significant reduction of diffusion for each elements especially Mg and calcium into the film. This wet-cleaning process also gives the best results on the
wettability of the substrate and a pure oxide surface after wet-cleaning, as well as the best results after annealing with a pure oxide surface and no change of optical properties of the substrate.

The choice of wet-cleaning therefore significantly affects the results observed by ToF-SIMS. We distinguished here three cases:

1) A well-defined interface with sharp interface zone (Al, ScMg⁺, and MgN⁻) for the sample DAE-F and AE-F.

2) A bad quality of the interface (a broad zone for Sc, Mg) due to diffusion and a also high diffusion of other elements such as as observed for the sample AI-F and HAE-F.

3) An intermediate case for the REF-F sample.

The quality of interface seems to be related to the amount of hydroxide and carbonates at the surface after annealing. Before film deposition, the sharpest interface with the lowest diffusion was found on the cleanest sample (DAE-F) and the worst interface was found on the AI-F sample with hydroxide and carbonates present after annealing. The decomposition of hydroxide and carbonates may continue during film deposition, which free the magnesium species and may cause the high diffusion of Mg, up to the surface of the film. This phenomenon was previously observed after the deposition of an oxide film on MgO(001), where magnesium was detected and migrated to the film surface of a 400 nm thick film. Following the same behavior, the initial low amount of calcium presence in the substrate may migrate into the film when using a high deposition temperature. Smilgys et al. demonstrated, using Auger spectroscopy, the migration of the calcium at the surface of the MgO(001) substrate after an annealing at 1100°C for 12h. In our study, even at 950°C, the diffusion of calcium occurred and migrated to the interface and into the film. The degree of diffusion into the film depends on the wet-cleaning process used.
The sample HAE-F is a special case here, with bad quality of the film/substrate interface and a high diffusion of Mg and Sc similar to the one observed on the sample AI-F. However, HAE-A was one of the samples with the lowest amount of hydroxides on the surface. The ToF-SIMS revealed a special feature at the interface with a plateau feature for all masses (even for Mg) and both in the positive and negative mode, which may indicate a change of chemistry or and effect of high roughness/porosity. Adding a sonication step in heptane removed the carbonates and hydroxides giving a pure oxide surface but, in the case of ScN film, it seemed to give a rough film/substrate interface. This degradation of film/substrate interface or surface of substrate could occur at the different steps of the experiment: cleaning, annealing and/or deposition.

The non-negligible presence of Mg in the AI-F and HAE-F film detected by $^{26}\text{Mg}^+$, $\text{MgN}^-$ or $\text{ScMg}^+$ may explain the presence of the observed secondary phase detected by XRD (figure 9). Also the presence of Sc extending into the MgO substrate is observed for the same samples and therefore can be present as an oxide $\text{ScO}_x$, also a possible explanation for the secondary phase seen in XRD. Therefore, both phases $\text{Sc}_2\text{O}_3$ and $\text{Mg}_3\text{N}_2$ may potentially be present at the film/substrate interface in both AI-F and HAE-F, as seen as a secondary phase in XRD.

The study of thin film deposition on MgO(100) substrates after different wet-cleaning processes, plus annealing revealed a significant difference at the film/substrate interface and film quality. The presence of hydroxides, carbonates at the surface prior to deposition seemed to increase the diffusion of Mg into the film up to the surface of the film (sample AI-F). The use of heptane combine with annealing at high temperature may degrade physically the surface of MgO(001) with an increase of the diffusion of Sc and Mg. The high diffusion of magnesium and scandium and calcium at the interface lead to the formation of a secondary phase such as $\text{Sc}_2\text{O}_3$ or $\text{Mg}_3\text{N}_3$ observed on the AI and HAE samples. The sample DAE-F and AE-F showed a minimum diffusion range/length with the sharpest film/substrate interface.
SUMMARY AND CONCLUSIONS

This paper studied the effect of different wet-cleaning processes on MgO(001) substrate monitored at different stage in thin film deposition. The surface chemistry of the substrate was checked after the different wet-cleaning processes, after in-situ annealing at 700°C for 2 h and after ScN thin film deposition with the study of the interface.

The as received substrate surface was contaminated. Sonication in solvents eliminate the main contaminants, but hydroxide and carbonates remains at the surface. The introduction of a sonication in a detergent before the one in acetone followed by ethanol lead to 100 % of MgO at the surface, i.e. recovery of the pure surface. The in-situ annealing was efficient for most of the different wet-cleaning even for the one without any wet-cleaning. Annealing the substrate removed most of the carbon contamination and lead to a surface composed at a minimum 80 % of MgO. The study of the interface ScN/MgO(001) revealed a significant difference in the diffusion of magnesium and scandium. The sharpest interface and lowest diffusion was observed for the sample cleaned by detergent followed by solvents.

In this study, two cleaning processes can be excluded for the MgO substrate. The first one is acetone followed by isopropanol which gave a contaminated surface with an interface characterized by broad diffusion range between the film and the substrate leading to the formation of a secondary phase. The second one is the use of heptane followed by acetone and ethanol which with the combination of annealing at 700°C/2h and a deposition of ScN at higher temperature may give a degraded surface or interface with a broad diffusion of element at the interface and the formation of a secondary phase in the film.

In conclusion the recommended cleaning process depend on the requirements for the film deposition and the applications. Nevertheless, the cleaning process using the sonication in detergent
followed by sonication in acetone and ethanol led to the cleanest surface with a pure oxide after wet-cleaning. This process is also the only one exhibiting a pure oxide surface after annealing while the other process led to a surface at the cleanest with a monolayer of hydroxide at the surface. Finally, among the different cleaning processes, the sonication in detergent followed by sonication in solvents led to the sharpest film/substrate interface in the case of ScN thin film deposition.

ACKNOWLEDGMENTS

The authors acknowledge Dr. G. Greczynski for XPS measurements and discussions. The authors acknowledge support from the European Research Council under the European Community’s Seventh Framework Programme (FP/2007-2013) / ERC grant agreement no 335383, the Knut and Alice Wallenberg Foundation through the Wallenberg Academy Fellows program, the Swedish Research Council (VR) through project grant no. 621-2012-4430, the Swedish Foundation for Strategic Research (SSF) through the Future Research Leaders 5 program, and the Swedish Government Strategic Research Area in Materials Science on Functional Materials at Linköping University (Faculty Grant SFO-Mat-LiU No 2009-00971).
APPENDIX: XPS FITTING DETAILS

Peak fitting was performed for all photoelectron peaks after a Shirley background subtraction and as described as follow using Voigt function for all components:

**Mg2s peak**

1. The mathematical function is a Voigt function with a fixed shape defined in KolXPD by $\text{FWHM}_{\text{gaussian}}/\text{FWHM}_{\text{lorentzian}} = 1.30$.
2. The full width at half maximum (FWHM) of identified peak component were kept fixed for all fitting: Mg-O (1.48 eV), Mg-OH$_2$ (1.50 eV), Mg-CO$_3$ (1.58 eV).
3. The energy separation between the different components were kept constant between the oxide and the hydroxide (+0.70 eV), the carbonate (+1.52 eV) components.

**O1s peak**

1. The mathematical function is a Voigt function with a fixed shape for each component and defined by $\text{FWHM}_{\text{gaussian}}/\text{FWHM}_{\text{lorentzian}}$ equal to 4.00 and 1.25 for the MgO and the contaminants component.
2. The full width at half maximum (FWHM) of the oxide component was kept fixed for all fitting: Mg-O (1.15 eV).
3. The energy separation between the oxide and the contaminant component (adsorbed species, gas) was kept constant at (2.20 eV) with a FWHM varying between 1.85 eV and 2.05 eV.
4. The hydroxide and carbonate component was impossible to separate and was deconvoluted from the other component as a common peak (Mg(OH)$_2$/MgCO$_3$) without constraints. The energy separation between the oxide and hydroxide/carbonate component varies from 0.55 to 0.80 eV and the FWHM varies from 1.40 to 1.62. The
shape \((\text{FWHM}_{\text{gaussian}}/\text{FWHM}_{\text{lorentzian}})\) varied from 1.50 to 4.00 when the ratio \(\text{MgCO}_3/\text{Mg(OH)}_2\) decrease from 0.6 to 0.
References


J. Libra.


TABLE I: a non-exhaustive list of the most used wet- and dry-cleaning for the thin film deposition and their effect on the substrate $^{2,5}$. (○ partially, possibly, ● completely)

<table>
<thead>
<tr>
<th>Purpose / effect</th>
<th>Wet-cleaning</th>
<th>Dry-cleaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>Remove of the dust, grease, lubricant</td>
<td>“Soap”, Surfactant, detergent, …</td>
<td>UV-ozone (soft etching)$^{11}$</td>
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<tr>
<td>Remove of organic contaminants</td>
<td>Commonly used solvents: acetone, methanol, ethanol, isopropanol</td>
<td>Plasma ion etching (ex-situ or in-situ)$^{12,14}$</td>
</tr>
<tr>
<td>Etching of the surface</td>
<td>Other solvents: n-heptane, trichloroethylene$^{6,7}$</td>
<td>Annealing (ex-situ or in-situ)$^{9,10,15,17}$</td>
</tr>
<tr>
<td>Reconstruction of the surface</td>
<td>Acid (H$_3$PO$_4$, H$_2$SO$_4$, HF) $^{8,10}$</td>
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<tr>
<td>Complex microstructure / patterning</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Expensive/complex technique</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Damage of the surface</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Need of post-processing</td>
<td></td>
<td></td>
</tr>
<tr>
<td>High risk assessment (environmental, health)</td>
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</table>
### TABLE 2: List of the different abbreviations used for the 18 samples studied depending of the wet-cleaning procedures, dry cleaning, and the deposition of ScN film.

<table>
<thead>
<tr>
<th>Wet-cleaning process</th>
<th>Name/abbreviation of the samples</th>
<th>Wet-cleaning</th>
<th>Wet-cleaning + annealing 700°C/2h</th>
<th>Wet-cleaning + annealing 700°C/2h + ScN thin film</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-received</td>
<td>REF</td>
<td>REF</td>
<td>REF-A</td>
<td>REF-F</td>
</tr>
<tr>
<td>Acetone + isopropanol</td>
<td>AI</td>
<td>AI-A</td>
<td>AI-F</td>
<td></td>
</tr>
<tr>
<td>Acetone + ethanol</td>
<td>AE</td>
<td>AE-A</td>
<td>AE-F</td>
<td></td>
</tr>
<tr>
<td>Heptane + acetone + ethanol</td>
<td>HAE</td>
<td>HAE-A</td>
<td>HAE-F</td>
<td></td>
</tr>
<tr>
<td>Detergent</td>
<td>D</td>
<td>D-A</td>
<td>DAE</td>
<td></td>
</tr>
<tr>
<td>Detergent + acetone + ethanol</td>
<td>DAE</td>
<td>DAE-A</td>
<td>DAE-F</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE 3: XRD analysis details of the (001)-ScN thin film deposited on MgO(001) substrate after the different cleaning processes: the cell parameter along the growth direction and the FWHM of the rocking curve/ω-scan of 002 ScN and 002 MgO and of the ϕ-scan performed on the{022}-ScN and {022}-MgO.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>a⊥(ScN) (Å)</th>
<th>ScN film ω-scan</th>
<th>ScN film ϕ-scan</th>
<th>MgO substrate ω-scan</th>
<th>MgO substrate ϕ-scan</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>FWHM 002-ScN</td>
<td>FWHM 022-ScN</td>
<td>FWHM 002-MgO</td>
<td>FWHM 022-MgO</td>
</tr>
<tr>
<td>REF-F</td>
<td>4.530(4)</td>
<td>0.79(2)°</td>
<td>1.07(2)°</td>
<td>0.04(2)°</td>
<td>0.06(2)°</td>
</tr>
<tr>
<td>AI-F</td>
<td>4.511(4)</td>
<td>1.32(2)°</td>
<td>2.02(2)°</td>
<td>0.05(2)°</td>
<td>0.06(2)°</td>
</tr>
<tr>
<td>AE-F</td>
<td>4.528(4)</td>
<td>0.82(2)°</td>
<td>1.05(2)°</td>
<td>0.05(2)°</td>
<td>0.07(2)°</td>
</tr>
<tr>
<td>HAE-F</td>
<td>4.529(4)</td>
<td>0.88(2)°</td>
<td>1.21(2)°</td>
<td>0.13(2)°</td>
<td>0.14(2)°</td>
</tr>
<tr>
<td>DAE-F</td>
<td>4.532(4)</td>
<td>0.81(2)°</td>
<td>1.05(2)°</td>
<td>0.04(2)°</td>
<td>0.06(2)°</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

FIG 1: photographs of the side view of a drop of water on the surface of MgO(001) substrate. Contact angles of deionized water drop for the a) the REF, b) the AI, c) the AE, d) the HAE, e) the D and f) the DAE samples.

FIG 2 (color online): XPS of the Mg2s peak of the surface of MgO substrate after wet-cleaning: a) the REF, b) the AI, c) the AE, d) the HAE, e) the D and f) the DAE samples. g) Composition of the surface of MgO substrate (estimated (±1%) from the fit on Mg2s).

FIG 3 (color online): XPS of the O1s peak of the surface of MgO substrate after wet-cleaning for a) the REF-A, b) the AI-A, c) the AE-A, d) the HAE-A, e) the D-A and f) the DAE-A samples. g) Composition of the surface of MgO substrate (estimated (±1%) from the fit on O1s after subtracting the contamination peak which represent an average of 20% of the total area).

FIG 4 (color online): XPS of the a) C1s peak of the surface of MgO substrate after wet-cleaning and b) the area of the C1s peak of the REF, the AI, the AE, the HAE, the D and the DAE samples.

FIG 5 (color online): Photographs of the MgO(001) substrates of the as-received (REF) substrate and after the in-situ annealing of the REF-A, the AI-A, the AE-A, the HAE-A, the D-A and the DAE-A samples.

FIG 6 (color online): XPS of the Mg2s peak of the surface of MgO substrate after wet-cleaning + annealing at 700°C/2h; for a) the REF-A, b) the AI-A, c) the AE-A, d) the HAE-A, e) the D-A and f) the DAE-A samples. g) Composition of the surface of MgO substrate (estimated (±1%) from the fit on Mg2s).

FIG 7 (color online): XPS of the O1s peak of the surface of MgO substrate after wet-cleaning + annealing at 700°C/2h; for a) the REF-A, b) the AI-A, c) the AE-A, d) the HAE-A, e) the D-A and f) the DAE-A samples. g) Composition of the surface of MgO substrate (estimated (±1%) from the fit on O1s after subtracting the contamination peak which represent an average of 20% of the total area).

FIG 8 (color online): XPS spectra of the C1s peak of the surface of MgO substrate after wet-cleaning + annealing at 700°C/2h: the REF-F, the AI-F, the AE-F, the HAE-F and the DAE-F samples.

FIG 9 (color online): XRD patterns of the ScN thin film deposited on MgO(001) after different wet-cleaning processes + in-situ annealing 700°C/2h: the REF-F, the AI-F, the AE-F, the HAE-F and the DAE-F samples. * indicates the secondary phase.

FIG 10 (color online): ToF-SIMS depth profile measured in the positive (right) and negative (left) mode for five ScN thin film deposited on MgO(001) using different wet-cleaning processes + in-situ annealing 700°C/2h prior to deposition: the REF-F, the AI-F, the AE-F, the HAE-F and the DAE-F samples.
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FIG 3 (color online): XPS of the O1s peak of the surface of MgO substrate after wet-cleaning for a) the REF-A, b) the Al-A, c) the AE-A, d) the HAE-A, e) the D-A and f) the DAE-A samples. g) Composition of the surface of MgO substrate (estimated (±1%) from the fit on O1s after subtracting the contamination peak contribution which, depending on samples, varies from 18 to 27% of the total area).
FIG 4 (color online): XPS of the a) C1s peak of the surface of MgO substrate after wet-cleaning and b) the area of the C1s peak of the REF, the AI, the AE, the HAE, the D and the DAE samples.

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FIG 8 (color online): XPS spectra of the C1s peak of the surface of MgO substrate after wet-cleaning + annealing at 700°C/2h: the REF-A, the AI-A, the AE-A, the HAE-A, the D-A and the DAE-A samples.

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