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Martijn F. J. Vos, Bart Macco, Nick F. W. Thissen, Ageeth A. Bol, and W. M. M. (Erwin) Kessels

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Atomic layer deposition of molybdenum oxide from (N^tBu)_2(NMe_2)_2Mo and O_2 plasma

Martijn F. J. Vos,^a Bart Macco,^a Nick F. W. Thissen, Ageeth A. Bol, and W. M. M. (Erwin) Kessels^b

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Molybdenum trioxide (MoO_3) films have been deposited by atomic layer deposition using bis(tert-butylimido)-bis(dimethylamido)molybdenum and oxygen plasma, within a temperature range of 50–350°C. Amorphous film growth was observed between 50 and 200°C at a growth per cycle (GPC) around 0.80 Å. For deposition temperatures of 250°C and higher, a transition to polycrystalline growth was observed, accompanied by an increase in GPC up to 1.88 Å. For all deposition temperatures the O/Mo ratio was found to be just below three, indicating the films were slightly substoichiometric with respect to MoO_3 and contained oxygen vacancies. The high purity of the films was demonstrated in the absence of detectable C and N contamination in Rutherford backscattering measurements, and a H content varying between 3 and 11 at. % measured with elastic recoil detection. In addition to the chemical composition, the optical properties are reported as well. © 2015 American Vacuum Society. [http://dx.doi.org/10.1116/1.4930161]

I. INTRODUCTION

Molybdenum trioxide has received considerable interest due to its optical, electrical, and catalytic properties. 1–4 Thin films of MoO_3 are being used in gas sensors, 5, 6 solid state lithium batteries, 7 and in the synthesis of MoS_2 by sulfurization. 8 Moreover, recently, MoO_3 has been used in organic photovoltaics, 9 perovskite solar cells, 10 and silicon solar cells as hole extraction layer. 11–15

A wide range of deposition techniques has been employed for the growth of MoO_3 films, such as thermal evaporation, 14 electron beam evaporation, 15 sputtering, 16–18 chemical vapor deposition (CVD), 19 and atomic layer deposition (ALD). 2, 20 Of these techniques, ALD offers the advantages of uniform and conformal growth, in conjunction with atomic level thickness control, merits which are highly desirable for some of the abovementioned applications. However, only few reports on ALD of MoO_3 can be found in literature. Diskus et al. deposited MoO_3 by ALD using molybdenum hexacarbonyl, Mo(CO)_6, as precursor and simultaneous doses of ozone and water as reactants. 20 However, the thermal stability of the Mo(CO)_6 precursor limited the ALD window to 152–172°C. Another study used MoF_6 and Si_2H_6 for the ALD of molybdenum films, which were subsequently post-treated with UV/ozone to obtain fully oxidized MoO_3. 9 However, the fact that this is a two-step process that involves the use of a halide precursor makes this process less ideal. More recently, Bertuch et al. reported an ALD process to deposit MoO_3 using bis(tert-butylimido)-bis(dimethylamido)-molybdenum, (N^tBu)_2(NMe_2)_2Mo, as the metal-organic precursor and ozone as reactant. 21 This process promisingly shows a high growth per cycle (GPC) of ~1.3 Å at 300°C, but suffers from a low GPC and from C and N contamination in the films at lower deposition temperatures of 150–200°C. Previously, the (N^tBu)_2(NMe_2)_2Mo precursor has also been used by Miikkulainen et al. to grow films of molybdenum nitride using ammonia as reactant. 22

In this study, we report on an ALD process to deposit high-purity substoichiometric molybdenum trioxide (MoO_3−x) films using the (N^tBu)_2(NMe_2)_2Mo precursor and O_2 plasma, for a wide temperature range of 50–350°C. O_2 plasma can offer the advantage of reasonable growth rates at lower deposition temperatures. In recent work, we have already shown the potential of this process for the low-temperature deposition of selective hole contacts for silicon heterojunction solar cells. 23 Here, we extend the temperature range to 350°C and provide a detailed study of the ALD process and material properties in general.

II. EXPERIMENT

A. Film deposition

MoO_3 films were deposited in a home-built ALD reactor, consisting of a vacuum chamber connected to an inductively coupled plasma source and a pump unit. The pump unit consists of a rotary and turbomolecular pump reaching a base pressure of ~10^{-6} Torr. An extensive description of the reactor can be found in earlier work from our group. 24 The typical processing conditions and experimental settings are summarized in Table I. The set temperature of the substrate table was varied between 50 and 350°C. The temperature of the reactor wall was maintained at 100°C, for all depositions, except for depositions at a table temperature of 50°C, for which the wall temperature was set to 50°C. The liquid (N^tBu)_2(NMe_2)_2Mo precursor (98%, Strem Chemicals) was contained in a bubbler at 50°C, at which it is reported to have a vapor pressure of 0.13 Torr. 21 The chemical structure of the precursor can be seen in the inset of Fig. I. The precursor supply line was heated to 80°C to prevent condensation of the precursor and...
Ar was used as a carrier gas. All depositions were done on Si (100) wafers with native oxide.

The standard ALD recipe consists of precursor dosing for 6 s in the first half of the ALD cycle. Ar gas is used as a carrier gas during the precursor dose, resulting in a chamber pressure of 7.5 mTorr. Subsequently, the reactor is purged with Ar for 3 s and pumped down for 3 s. The second half-cycle consists of O₂ plasma exposure with a plasma power of 6 s in the first half of the ALD cycle. Ar gas is used as a carrier gas during the precursor dose, resulting in a chamber pressure 5.1 mTorr.

### TABLE I. Standard ALD recipe for the deposition of MoOₓ.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deposition temperature</td>
<td>50–350°C</td>
</tr>
<tr>
<td>Wall temperature</td>
<td>50–100°C</td>
</tr>
<tr>
<td>Bubbler temperature</td>
<td>50°C</td>
</tr>
<tr>
<td>Precursor line temperature</td>
<td>80°C</td>
</tr>
<tr>
<td>Base pressure</td>
<td>10⁻⁶ Torr</td>
</tr>
<tr>
<td>Ar pressure</td>
<td>7.5 mTorr</td>
</tr>
<tr>
<td>O₂ pressure</td>
<td>5.1 mTorr</td>
</tr>
<tr>
<td>Precursor dosing time</td>
<td>6.0 s</td>
</tr>
<tr>
<td>Precursor purge time</td>
<td>3.0 s</td>
</tr>
<tr>
<td>Plasma exposure time</td>
<td>4.0 s</td>
</tr>
<tr>
<td>Reactant purge time</td>
<td>3.0 s</td>
</tr>
</tbody>
</table>

B. Film analysis

The growth of the MoOₓ films was monitored *in situ* by spectroscopic ellipsometry (SE) using a J.A. Woollam, Inc., M2000U ellipsometer. The dielectric function of the deposited films was parameterized using a combination of a Tauc-Lorentz and a Gaussian oscillator, as will be addressed later in this paper.

X-ray photoelectron spectroscopy (XPS) was done with a Thermo Scientific KA1066 spectrometer, using monochromatic Al Kα x-rays with an energy of 1486.6 eV. Rutherford backscattering spectroscopy (RBS) and elastic recoil detection (ERD) were performed by AccTec BV, Eindhoven, The Netherlands, using a Singleton with a 2 MeV He⁺ beam to determine the chemical composition of the films. The mass density was calculated using the areal mass density as obtained from RBS/ERD measurements and the film thickness as obtained from SE. Raman spectroscopy measurements were done using a Renishaw Invia Raman microscope, using a laser wavelength of 514 nm. Grazing incidence x-ray diffraction (XRD) measurements were done with a PANalytical X’Pert Pro MRD system, using Cu Kα x-rays (λ = 1.54 Å). Scanning electron microscopy (SEM) images were obtained using a FEI Nova600i NanoLab.

### III. RESULTS AND DISCUSSION

A. ALD growth

To study the ALD process as a function of the deposition temperature, a set of approximately 20 nm thick samples was deposited at temperatures between 50 and 350°C, using a standard saturated precursor dosing time of 6 s and a plasma exposure time of 4 s. In Fig. 1, the film thickness is shown as a function of ALD cycles for temperatures between 50 and 350°C. The thickness of the deposited films proceeds linearly with the number of ALD cycles for deposition temperatures of 50 and 200°C, corresponding to a GPC of ~0.8 Å. For temperatures of 300 and 350°C a gradual increase in GPC during film growth can be observed, which is attributed to a transition from amorphous to polycrystalline growth. Such a transition has been observed for other ALD processes before, for instance, for the deposition of TiO₂.[25,26] A transition from amorphous to polycrystalline growth is accompanied by the appearance of grains, very likely leading to an increase in surface area and therefore to an increase in GPC. This will be further corroborated in Sec. III B 2, where the crystallinity and morphology of the films will be discussed.[27] Note that in addition to the increased roughness, an enhanced reactivity of the polycrystalline surface can also contribute to the increase in GPC. To exclude the presence of a thermal CVD component as a possible explanation for the enhanced growth rate at higher deposition temperatures, it was verified that no growth occurs when using O₂ gas instead of O₂ plasma as reactant.

In Fig. 2, the saturation curves for the precursor dose step [Fig. 2(a)] and plasma exposure step [Fig. 2(b)] are shown for various temperatures. The GPC as a function of precursor dosing time [Fig. 2(a)] shows a relatively soft saturation and a precursor dosing time of 6 s was chosen as standard for all deposition temperatures. The plasma saturation curve [Fig. 2(b)] shows a slightly higher GPC for low plasma exposure times, likely corresponding to incomplete combustion of precursor ligands or redeposition.[28] To assure complete combustion of ligands and to prevent impurity incorporation in...
the films a plasma exposure time of 4 s was chosen as standard.

In Fig. 3, the GPC as a function of deposition temperature is shown, both in terms of thickness, determined from SE, and in terms of deposited Mo atoms/nm², determined from RBS. Note that the samples used for determination of the GPC data in Fig. 3 correspond to the samples used in Fig. 1 and had a thickness of ~20 nm. As can be seen in Fig. 3, the GPC determined by SE (closed squares) varies only slightly for temperatures between 50 and 200 °C. At these deposition temperatures, the films are completely amorphous (see Sec. III B 2). For higher temperatures, the GPC increases, which is due to a transition to polycrystalline growth. For a deposition temperature of 300 °C, a distinction can be made between amorphous growth at the start of the deposition, and polycrystalline growth at a larger film thickness, as is also shown in Fig. 3. The GPC determined from the first 20–100 cycles was found to be 0.93 Å, while calculating the GPC from the last 50 cycles of the deposition at 300 °C yields a GPC of 1.25 Å. Note that these values for the GPC were calculated by determining the slope at different positions on the curves in Fig. 1. Since at 350 °C the amorphous-polycrystalline transition occurs already at a very early stage, only the GPC for polycrystalline growth is reported for this temperature.

Figure 3 also shows the GPC in terms of the number of deposited Mo atoms/nm² (open circles), as determined from RBS, which shows an increase with temperature. Note that this value was determined by dividing the total number of Mo atoms/nm² by the number of ALD cycles. The value is therefore an average over all the ALD cycles used to deposit the film. For this reason this quantity is denoted as \( \langle \text{GPC}_{\text{RBS}} \rangle \). For a direct comparison of the RBS and SE data, also the average GPC as determined from SE (\( \langle \text{GPC}_{\text{SE}} \rangle \)), which is the final film thickness divided by the number of cycles, is shown in Fig. 3 (open squares). As can be seen, there is a very strong correlation between \( \langle \text{GPC}_{\text{RBS}} \rangle \) and \( \langle \text{GPC}_{\text{SE}} \rangle \), which demonstrates that the increased GPC at high temperatures is mainly caused by an actual increase in the number of deposited Mo atoms per cycle and not by a decrease in film density. This increase in GPC at high temperatures is thought to be caused by an increase in surface area due to increased roughness, an increased reactivity of the surface, or a combination of both.²⁷

### B. Film characterization

In this section, the chemical composition, morphology, and optical properties of the ~20 nm films deposited at temperatures between 50 and 350 °C will be discussed. The most important film properties have been summarized in Table II.

#### 1. Chemical composition

From RBS/ERD measurements, both the stoichiometry and the elemental composition were determined. The O/Mo ratio and the atomic percentage of H as a function of...
deposition temperature can be found in Table II. The O/Mo ratio was found to be below 3 for all investigated deposition temperatures (2.9 on average), indicating the films were slightly substoichiometric with respect to molybdenum trioxide (O/Mo ratio = 3). Note that the samples were transferred in ambient and therefore an effect of ambient on the film stoichiometry cannot be excluded. For all deposition temperatures the contents of C and N impurities were below the detection limit of 3 and 2 at. %, respectively, showing the high quality of the films. No substantial C or N contamination is present in the bulk of the films, which is in line with the fact that there is no indication of thermal decomposition of the precursor molecule for high decomposition temperatures. The H content of the films deposited at 50 and 100 °C was found to be around 10 at. %, which decreased to ~5 at. % for higher temperatures. This H can originate from the ligands of the precursor molecule, which contains 30 H atoms, as well as from residual water in the reactor.

The mass density of the films was calculated using the RBS/ERD measurements and the layer thicknesses from SE. For deposition temperatures from 100 to 300 °C the density was found to be around 4.2 g cm\(^{-3}\), which is slightly lower than the bulk density of crystalline MoO\(_3\) of 4.69 g cm\(^{-3}\). The density is somewhat lower (3.9 g cm\(^{-3}\)) for 50 and 350 °C.

XPS was used to further study the chemical composition of the deposited films. In Fig. 4(a), the Mo3d peak is depicted for a deposition temperature of 50 and 350 °C. It was found that sputtering of MoO\(_x\) with Ar ions causes a change in stoichiometry of the material due to preferential O sputtering, which is known from literature.\(^{29}\) For this reason, the XPS data in Fig. 4 were collected prior to any sputtering, and thus, the presence of surface contamination cannot be excluded.

The Mo3d XPS spectrum in Fig. 4(a) shows a dominant doublet with the Mo3d\(_{5/2}\) peak at ~233.0 eV and the Mo3d\(_{3/2}\) peak at 236.2 eV, corresponding to the fully oxidized Mo\(^{6+}\) state, i.e., MoO\(_3\).\(^{29,30}\) The binding energy of the Mo3d\(_{5/2}\) peak is slightly higher than other literature reports, which is likely an effect of surface charging.\(^{17}\) A second doublet at 231.7 and 235.0 eV is required to obtain a good fit to the experimental data. This doublet has previously been attributed to the Mo\(^{5+}\) state, corresponding to oxygen vacancies.\(^{11,31}\) For instance, Battaglia et al. demonstrated an increase in the Mo\(^{5+}\) peak upon annealing the samples in N\(_2\) environment, whereas no change was observed when annealing in O\(_2\) environment.\(^{11}\) Similar experiments were carried out in this study and yielded similar results, showing an increase in the Mo\(^{5+}\) state and even the appearance of the Mo\(^{4+}\) state upon annealing at 500 °C in N\(_2\) environment (not shown here).

The presence of oxygen vacancies is also consistent with the

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**Table II. Properties of MoO\(_x\)**

<table>
<thead>
<tr>
<th>Deposition temperature (°C)</th>
<th>GPC (Å)</th>
<th>Mo (at. nm(^{-2}) cycle(^{-1}))</th>
<th>O/Mo</th>
<th>[H] (at. %)</th>
<th>Mass density (g cm(^{-3}))</th>
<th>Refractive index (at 1.96 eV)</th>
<th>(E_g) (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>0.83 ± 0.03</td>
<td>1.41 ± 0.05</td>
<td>2.9 ± 0.1</td>
<td>11 ± 1</td>
<td>3.9 ± 0.2</td>
<td>2.11 ± 0.03</td>
<td>2.87 ± 0.03</td>
</tr>
<tr>
<td>100</td>
<td>0.72</td>
<td>1.42</td>
<td>2.8</td>
<td>10</td>
<td>4.3</td>
<td>2.20</td>
<td>2.80</td>
</tr>
<tr>
<td>150</td>
<td>0.74</td>
<td>1.37</td>
<td>2.9</td>
<td>3.3 ± 0.4</td>
<td>4.2</td>
<td>2.19</td>
<td>2.81</td>
</tr>
<tr>
<td>200</td>
<td>0.80</td>
<td>1.47</td>
<td>2.9</td>
<td>3.3</td>
<td>4.2</td>
<td>2.18</td>
<td>2.80</td>
</tr>
<tr>
<td>250</td>
<td>0.93</td>
<td>1.52</td>
<td>3.0</td>
<td>3.6</td>
<td>4.2</td>
<td>2.13</td>
<td>2.77</td>
</tr>
<tr>
<td>300</td>
<td>1.25</td>
<td>1.84</td>
<td>2.9</td>
<td>4.3</td>
<td>4.2</td>
<td>2.12</td>
<td>2.92</td>
</tr>
<tr>
<td>350</td>
<td>1.88</td>
<td>2.48</td>
<td>2.9</td>
<td>4.1</td>
<td>3.9</td>
<td>1.98</td>
<td>2.92</td>
</tr>
</tbody>
</table>
substoichiometric O/Mo ratio as was found with RBS. The O1s peak in Fig. 4(b) consists of a dominant peak at approximately 530.9 eV, corresponding to O bound to Mo atoms\textsuperscript{29} and a small shoulder around 532.5 eV, which could possibly be explained by O–H bonds\textsuperscript{31} or by absorbed surface species.\textsuperscript{17} Although the presence of C and N was observed in the XPS spectrum, their signals were almost completely absent after shortly sputtering with Ar\textsuperscript{+} ions. This indicates that C and N were mainly present on the surface, which is consistent with the RBS results.

2. Crystallinity and surface morphology

Raman spectroscopy and XRD were used to assess the crystallinity of the films as a function of the deposition temperature. From Fig. 5(a) it can be seen that the film deposited at 250°C shows two peaks, at 777 and 849 cm\textsuperscript{-1}, corresponding to the metastable monoclinic β-crystal phase.\textsuperscript{32,33} For 300 and 350°C a clear peak is visible at 818 cm\textsuperscript{-1}, indicating the films are in the stable orthorhombic α-phase. The absence of these peaks for temperatures of 200°C and lower confirms their amorphous nature. The x-ray diffractograms in Fig. 5(b) show similar results of crystalline films at higher deposition temperatures. The (110) peak in Fig. 5(b) corresponds to the β-phase, while the (020), (040), and (060) peaks originate from the α-phase.\textsuperscript{34} For 300°C the Raman spectrum in Fig. 5(a) only shows the presence of the α-phase, while the x-ray diffractogram in Fig. 5(b) shows mainly the α-phase, with a small additional contribution originating from the β crystal-phase. For 200 and 250°C no peaks are visible, corresponding to amorphous material.

Although Raman spectroscopy shows crystallinity for a deposition temperature of 250°C it is thought that the bulk material is amorphous and some small crystallites are present at the surface of the film. This can also be seen in the SEM images in Fig. 6.

The film morphology as a function of deposition temperature was studied by top-view SEM (Fig. 6). For a deposition temperature of 50°C, a relatively smooth surface is observed, corresponding to amorphous film growth. At 250°C, some small nuclei are visible, surrounded by amorphous material. The film deposited at 300°C contains many, larger crystallites, but amorphous material is still visible in the background. For 350°C, the surface of the film appears fully crystalline. The appearance of crystallites and increasing crystallinity at higher temperatures is in agreement with the Raman data as presented in Fig. 5, as well as the observations made based on Fig. 1. Moreover, the increased roughness at higher deposition temperatures was also confirmed with atomic force microscopy (not shown), which showed a RMS roughness of 0.2 nm for 50°C vs 1.9 nm for 350°C.

3. Optical properties

The optical properties of the MoO\textsubscript{x} films have been analyzed using SE. A Tauc-Lorentz oscillator was used to account for interband absorption. Additionally, a Gaussian oscillator at low photon energy (∼1 eV) was used for absorption inside the band gap, likely caused by defects. This absorption at low photon energy has been reported earlier for MoO\textsubscript{x} and has been attributed to oxygen defects.\textsuperscript{4,11,16,35}
The refractive index \((n)\) and the extinction coefficient \((k)\) obtained from the modeling are shown in Fig. 7.

The refractive index in Fig. 7 lies in the 1.8–2.2 range, which is consistent with other literature on MoO\(_3\).\(^{16,36}\) Although the extinction coefficient \(k\) varies only slightly with deposition temperature, the refractive index \(n\) is strongly dependent on the deposition temperature, likely caused by differences in film density. From the absorption coefficient \(\alpha\) obtained from the SE measurements the Tauc band gap was determined by plotting \((\alpha h\nu)^{1/2}\) as a function of photon energy (considering MoO\(_3\) is an indirect band gap material) and extrapolating the linear part to zero absorption.\(^{37}\) The results of this procedure are listed in Table II. The obtained band gap values of 2.7–2.9 eV are in the range of the values of 2.5–3.0 eV previously reported in the literature.\(^ {1,2,36,38}\)

IV. SUMMARY AND CONCLUSIONS

An ALD process to deposit high-purity MoO\(_3\) films has been presented, which is based on \((\text{N}^{+}\text{Bu})_3(\text{NMMe}_2)_2\) Mo dosing and O\(_2\) plasma exposure and which yields a high GPC over a temperature range of 50–350°C. For amorphous growth at low temperature (<250°C), GPC values of 0.72–0.93 Å were obtained. For higher deposition temperatures, a transition to polycrystalline growth occurs, which is accompanied by an increase in GPC up to 1.88 Å for 350°C. It was shown that this increase in GPC is consistent with an increase in the amount of deposited Mo at nm\(^{-2}\) cycle\(^{-1}\).

For all deposition temperatures the MoO\(_3\) films were found to be slightly substoichiometric with respect to MoO\(_3\). Contamination by C and N was determined to be below the RBS detection limit of 3 and 2 at. %, respectively, which shows the high quality of the material. In addition, the MoO\(_x\) films contain 3–11 at. % hydrogen. The Tauc band gap was found to vary between 2.77 and 2.87 eV for amorphous films and was 2.92 eV for polycrystalline films deposited at 300 and 350°C.

Due to the high purity of the films and the relatively high GPC, the presented process is likely suitable for many applications, such as photovoltaics and other optoelectronic devices.

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