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## Sub-unit-cell control of LaAlO3/SrTiO3 interfaces near the critical thickness

--Manuscript Draft--

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| **Abstract:** | The two-dimensional electron gas (2DEG) at the interface between a polar (LaAlO3 or LAO) and non-polar (SrTiO3 or STO) insulating oxide offers opportunities to develop multifunctional devices. The prospect of this interface will depend on both its performance compared to existing technologies and its ability to integrate with CMOS technology. As a measure for the 2DEG electronic quality, the carrier density and mobility are commonly used, and they strongly relate to the quality of the hosting material and interface. A good understanding of the material growth and interface formation therefore would be required to be able to control and tune the electron density and mobility. Using pulsed laser deposition with in-situ reflection high energy electron diffraction (RHEED), we demonstrate the control of LAO growth at sub-unit-cell precision. While it is commonly observed that the LAO/STO shows a metal-to-insulator transition (MIT) at around 3 to 4 unit-cells (uc) of LAO, our results demonstrate that it occurs precisely at 3.65 uc (with the last LAO layer having only 65% coverage). Near this critical value, we further show that the electron mobility is sensitive to the degree of the surface coverage, carrier density (which can be varied by back gating) and magnetic scattering (which is seen at high carrier concentrations). In this paper, the sample fabrication and characterization process are elaborated and we propose that this process might also be applicable for growing other (oxide) interfaces. |
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The Editor

JoVE,

Dear Editor,

We would like to submit the manuscript, **Controlled fractional unit cell growth of $LaAlO_3/SrTiO_3$ interfaces near the critical thickness and its scattering properties** by C. J. Li et al., for consideration in JoVE.

The two dimensional electron gas (2DEG) generated at the $LaAlO_3/SrTiO_3$ **(**LAO/STO) interface has been a very exciting development over the last decade and the potential for oxide electronic devices based on these interfaces would require a clear understanding of the electronic transport properties at these interfaces and our ability to manipulate them at will.

For this purpose, we developed a standard protocol for controlled fractional unit cell growth of LAO, patterning the 2DEG and characterization of the patterned 2DEG. Using these techniques, we have successfully identified the exact critical thickness for the metal-insulator transition of 3.65 uc. Further, the carrier carrier scattering, strain and Kondo induced scattering are three major scattering mechanisms affecting the transport properties of LAO/STO interfaces. In this paper, we illustrate the detailed procedures of the experiments to make it available to wide research community and applicable to other oxide interface systems.

We feel techniques described in this paper are simple and general enough and will lead to a broad interest in general oxide thin film fabrication and characterization community and hence eminently suited for publication in JoVE.

Sincerely,

Changjian Li and T. Venkatesan.

**TITLE:**
**Sub-unit-cell control of $LaAlO_3/SrTiO_3$ interfaces near the critical thickness**

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**KEYWORDS:**
Aluminum Nitride (AlN) mask, photolithography, reflection high energy electron diffraction (RHEED), field effect, $LaAlO_3/SrTiO_3$ interface, metal-insulator transition, mobility, strain, Kondo effect

**SHORT ABSTRACT:**
We present a standard protocol for growing sub-unit-cell $LaAlO_3$ near the critical thickness of the $LaAlO_3/SrTiO_3$ metal-to-insulator transition using pulsed laser deposition with *in-situ* reflection high energy electron diffraction (RHEED). Electric field effect measurements of these sub-unit-cell interface samples reveal different scattering mechanisms.

**LONG ABSTRACT:**
The two-dimensional electron gas (2DEG) at the interface between a polar ($LaAlO_3$ or LAO) and non-polar ($SrTiO_3$ or STO) insulating oxide offers opportunities to develop multifunctional devices. The prospect of this interface will depend on both its performance compared to existing technologies and its ability to integrate with complementary metal-oxide-semiconductor (CMOS) technology. As a measure for the 2DEG electronic quality, the carrier density and mobility are commonly used, and they strongly relate to the quality of the hosting material and interface. A good understanding of the material growth and interface formation therefore would be required to be able to control and tune the electron density and mobility. Using pulsed laser deposition with in-situ reflection high energy electron diffraction (RHEED), we demonstrate the control of LAO growth at sub-unit-cell precision. While it is commonly observed that the LAO/STO shows a metal-to-insulator transition (MIT) at around 3 to 4 unit-cells (uc) of LAO, our results demonstrate that it occurs precisely at 3.65 uc (with the last LAO layer having only 65% coverage). Near this critical value, we further show that the electron mobility is sensitive to the degree of the surface coverage, carrier density (which can be varied by back gating) and magnetic scattering (which is seen at high carrier concentrations). In this paper, the sample fabrication and characterization processes are elaborated and we propose that these processes are also applicable for growing other (oxide) interfaces.

**INTRODUCTION:**

Since the first report by Ohtomo and Hwang[1], two-dimensional electron gas (2DEG) at the interface between a polar insulator $LaAlO_3$ (LAO) and non-polar insulator $SrTiO_3$ (STO) has attracted extensive research in order to understand its origin[2-5] and explore its properties[6-8] and potential applications.[9-12] To exploit the 2DEG in electronic devices, the ability to control and tune its electronic properties is a prerequisite. A good understanding on the 2DEG scattering mechanism is thus required, which directly relates to the interface carrier density and mobility.

It is generally observed that the insulator-to-metallic transition at the LAO/STO interface occurs when the LAO thickness is around 3-4 unit-cells (uc). However, the observed values of carrier density and mobility at the critical thickness tend to vary from one report to another, and this might be due to that the precise LAO thickness is less controlled. In previous studies[15], the top (last) LAO layer during deposition is usually assumed to be an integer of unit cells, i.e. a layer with a complete coverage. In this paper, we established a method to precisely control the growth of LAO layer with sub-unit-cell precision near the critical thickness of between 3 and 4 uc by monitoring the RHEED intensity oscillation during the LAO growth using PLD. In addition, we developed a technique to create micron patterns of the 2DEG based on lithographically patterned amorphous AlN layer before the growth of the LAO polar layer. The motivation is that a good recipe to chemically etch oxide layer is not yet established while the ion-beam etching technique may result in defects formation on the STO surface that may lead to an unwanted surface conductivity. Furthermore, an ion beam etching facility is not always available in every research lab. Another alternative is a lift-off process. However, as the deposition temperature to produce a crystalline LAO is relatively high (650 °C and above), a normal photoresist cannot withstand the high temperature. We have developed a process to pattern amorphous AlN as a replacement for the photoresist, and in this case a negative image of the desired pattern has to be produced. The use of AlN as a mask allows the subsequent deposition of LAO to be performed at high temperatures. The area uncovered by AlN defines the final pattern of our structures and the deposition of LAO directly on this area produces interface conductivity, while the LAO deposited on AlN remain insulating. This process allows patterning of the 2DEG without complicated and damaging etching process.

The carrier density and mobility of the samples were characterized using Hall effects, showing that the 2DEG starts to emerge when the LAO thickness is 3.65 uc (65% coverage of the fourth LAO layer) and the 2DEG mobility is sensitive to the surface coverage above this critical value. In order to understand the relation between carrier density and mobility, the 2DEG density was tuned using electric field with the STO substrate as the dielectric. The Hall and field effect data show that the mobility depends on the coverage of the top LAO layer and carrier density, indicating that it is governed by various scattering mechanisms including carrier-carrier, strain induced defect, and magnetic (only observed at high carrier concentration) scatterings. These results provide guidance in tuning the 2DEG and designing future oxide electronics.

**PROTOCOL:**

## 1. Substrate preparation

1.1 Obtain commercially available single- or double-side polished single-crystal STO (001) substrates of 0.5 mm thick and 5x5 $mm^2$ surface area.

1.2 Immerse and ultrasonically soak the as-received STO substrates in 0.1 L deionized water for 10 min. The topmost layer of SrO domains will form Sr-hydroxide complex, which can be dissolved in acidic solution.

1.3 Immediately dip the substrates into 50 mL buffered hydrofluoric acid (BHF) solution and ultrasonically soak for 30 s. Subsequently, rinse the substrates with deionized water and dry them with nitrogen spray. The BHF will selectively etch away the SrO domains, producing practically perfect $TiO_2$ termination on the STO surface.

1.4 To remove the remnants of the previous treatments and facilitate recrystallization, anneal the substrates in a furnace in an atmospheric pressure, with the following steps; heat up to 950 °C at a rate of 5 °C/min, keep at 950 °C for 1.5 hours, and cool down to room temperature at a rate of 3 °C/min.

1.5 Perform atomic force microscopy (AFM) in a tapping mode with a scanning rate of 1 Hz over an area of 3×3 $\mu m^2$ in order to characterize the surface morphology according to manufacturer's protocol.

1.5.1. Use the built-in AFM software to open and analyze the obtained image files. If necessary, subtract any background and measurement artifacts. A single $TiO_2$ terminated surface should show clear atomic terraces.

1.5.2. Determine the root mean square (rms) roughness of the surface and the width and height (should correspond to the lattice constant of STO of around 0.4 nm) of the terraces. Export the processed images into .jpg file. Figure 1 shows the evolution of the STO surface morphology captured after each treatment step.

## 2. Photolithography

2.1 Load and secure a BHF treated STO substrate onto a spin coater, coat with 0.5 mL poly(methyl methacrylate) (PMMA) photoresist, and immediately spin at 3000 rpm for 1 min. Unload the substrate from the spin coater and bake it on a hot plate at 120 °C for 1 min. Repeat this procedure for all other substrates.

2.2 Load and secure the substrate in the UV mask aligner. Align the pre-designed mask (with Hall bar structures) on the sample and expose for 8 s. The Hall bar mask is manufactured using laser writing process with the bar dimension of 160 µm in length and 100 µm in width (see Figure 2a). Repeat this procedure for all other substrates.

2.3 Unload the substrate from the mask aligner and immerse in PMMA developer for 1 min. The positive image of the Hall bar will be obtained on the surface. Repeat this procedure for all

other substrates.

## 3. AlN sacrificial mask deposition

3.1 Secure the aluminum nitride (AlN) target on a Pulse Laser Deposition (PLD) target holder, and glue the backside of one of the STO on a substrate holder using ~2 mL silver paint and place the holder on a hot plate for ~10 min at 100 °C to dry the silver paint and allow a good thermal contact between the holder and substrate. Load both the target and substrate holders into the PLD deposition chamber.

3.2 Adjust the substrate-target distance to 7 cm and wait for the base pressure of the PLD chamber to reach at least $1 \times 10^{-7}$ Torr.

3.3 After the expected base pressure is obtained, introduce pure oxygen into the deposition chamber and adjust its flow until a stable pressure of ~$1 \times 10^{-6}$ Torr is achieved.

3.4 Align the Laser optics, adjust the Laser power and to obtain an energy fluence of 1.5 J/cm$^2$ on the target surface, and set the repetition rate to 5 Hz. Rotate and raster the target during laser ablation to uniformly ablate the target surface area.

3.5 Start a predeposition by ablating the AlN target with the laser with the target shutter closed to clean the AlN target. After 5 min predeposition, open the target shutter to start the deposition.

3.5.1. To produce a uniform layer, rotate the substrate during laser ablation. After a 30 min deposition to obtain 100 nm thick AlN, stop the deposition by switching-off the laser. The growth rate of AlN has been previously calibrated. An amorphous AlN layer is produced and it fully covers the previously structured photoresist.

3.6 Stop all target and substrate rastering and rotation, and switch-off the oxygen flow. Unload the substrate from the chamber.

3.7 Immerse the substrate into a beaker filled with ~50 mL acetone and ultrasonically soak for 1 min. Rinse the substrate with deionized water and make sure photoresist is completely removed.

3.8 Observe the obtained Hall bar pattern under an optical microscope; a representative pattern is shown in Figure 2a. Repeat the cleaning process if necessary.

## 4. LAO Film deposition

4.1 Using the same process as 3.1, load a LAO single crystal target and the STO substrate obtained from section 3 into the PLD chamber.

4.2 Wait for the base pressure of the chamber to reach $1 \times 10^{-7}$ Torr.

4.3 Introduce pure oxygen into the deposition chamber and adjust its flow until a stable pressure of $1 \times 10^{-3}$ Torr is achieved.

4.4 Turn on the RHEED electron gun and set the voltage to 25 kV and filament current to 1.5 A. Open the valve between the RHEED and deposition chamber. Turn on the RHEED CCD camera.

4.5 Adjust the z-position and rotation-angle of the substrate with respect to the electron beam until a good RHEED pattern is observed on the CCD camera. An example of a good RHEED pattern is shown in Figure 3 for different steps for thin film growth. The z-position is ~86 and the rotation angle is not fixed.

4.6 Heat up the substrate heater at 15 °C/min until it reaches and stabilizes at 900 °C (corresponding to a substrate temperature of about 750 °C). Wait for about 5 minutes.

4.7 Repeat step 3.4 and set Laser fluence at 1.5 J/cm$^2$ and repetition rate at 2 Hz, and perform predeposition for 5 mins. Meanwhile, observe the RHEED pattern and, if necessary, adjust again the sample position to get a good RHEED pattern (see Figure 3).

4.8 Start deposition by opening the target shutter. Using the RHEED software, monitor the intensity of the specular spot (specified in Figure 3) of the image on the CCD camera. The software will display and record a real time evolution of the spot intensity, showing a periodic oscillation. Figure 4c and 4d show examples of the intensity oscillation.

4.9 Determine the periodicity of the oscillation by counting the peak-to-peak duration. One period corresponds to one unit-cell (~0.4 nm) growth of LAO. The growth rate in our chamber with the deposition parameters described above is 0.025 uc/sec.

4.10 Based on the growth rate, stop the laser at the desired LAO thickness. For example, to obtain 3.50 uc samples, stop the laser at 140 sec mark. Figure 4 shows an example of the sub-unit-cell growth control process.

4.11 Stop all target and substrate rastering and rotation, and set the substrate temperature to 25 °C at 10 °C/min while keeping the pressure the same as the deposition pressure.

4.12 When the sample temperature reaches 100 °C or lower, switch off the oxygen flow. Unload the sample from the chamber.

4.13 Anneal the sample by heating it up to 600 °C at 10 °C/min in a furnace with an oxygen flow at 0.5 bar. Keep the sample in the furnace for 1 hour and subsequently cool it down to room temperature at 10 °C/min. This step removes oxygen vacancies from the sample and minimizes their contribution to the conductivity.

4.14 Repeat steps 4.1 to 4.13 for deposition of different LAO thicknesses (3.5, 3.65, 3.8 and 4 uc).

**5. Atomic force microscopy**

5.1 Load the sample onto the AFM sample stage.

5.2 Load the AFM cantilever and make an adjustment to the laser and photodiode suitable for scanning in a tapping mode.

5.3 Scan the sample surface with a scanning rate of 1 Hz and scanning area of $3 \times 3\ \mu m^2$.

5.4 Analyze the AFM images with the same method discussed in section 1.5. Some obtained images are shown in Figure 4.

5.5 Repeat steps 5.1 to 5.4 for other samples with different thicknesses (3.5, 3.65, 3.8 and 4 uc).

**6. Hall and electric field effects**

6.1 Paste a sample on a copper plate by uniformly covering the backside of the sample with approximately 2 mL silver paint. This ensures a good electrical contact between the copper plate and the substrate and allows electric fields to be applied to the STO substrate through the copper plate for the field effect measurement.

6.2 Using approximately 2 mL low temperature grease, secure the copper plate on a sample holder suitable for the physical properties measurement system (PPMS) with the sample surface facing up.

6.3 Form electrical connections between the sample and sample holder by Al wire for each respective electrical pads using ultrasonic wire bonder with a bonding power of 270 W and duration of 30 msec. The connection layout for the samples is shown in Figure 2b.

6.4 Load the sample holder into the PPMS chamber and stabilize the temperature by inputting a set point of 300 K. Set the measurement (driving) current to 1 µA and the potential range of the source meter for the gate voltage to 50 V.

6.5 Switch on the applied magnetic field and sweep the field from 0 to 9 T while measuring the longitudinal or perpendicular (Hall) resistance through their respective potential connections. (Figure 2b)

6.6 Observe any leakage current through the bottom STO gate through a multimeter during application of a gate voltage. The leakage current must be less than 10 nA.

6.7 Repeat step 6.4 for different applied gate voltages (from -50 V to 50 V with a 10 V step) and for different temperatures (300, 200, 100, 50, 20, 10, 5 and 2 K).

6.8 Repeat steps 6.1 to 6.6 for other samples with different thicknesses (3.5, 3.65, 3.8 and 4 uc).

6.9 Analyze the resistance-magnetic field dependence curves and calculate the carrier density and mobility of the samples by using the following relations:

$n = -\frac{1}{e \cdot Hall\ slope}$ (1)

$\mu = \frac{L}{RW} \cdot \frac{1}{ne} = \frac{L}{RW} \cdot (-Hall\ slope)$ (2),

where n and μ are carrier density and mobility, respectively, R is the linear resistance, *Hall slope* is the gradient of the Hall resistance versus magnetic field, L and W are the length (160 μm) and the width (100 μm) of the Hall bar, respectively, and e is the elementary charge.

The magnetoresistance (MR) is defined as MR(B)= R(B)-R(0)/R(0) × 100 %, where R(B) is the resistance at an applied magnetic field B and R(0) is the resistance at zero magnetic field.

**REPRESENTATIVE RESULTS:**

The substrate preparation process greatly affects the quality of STO surfaces. Figure 1 shows the evolution of the surface morphology before and after BHF and thermal treatment. The as-received substrates show a very smooth surface without atomic terraces. After BHF treatment, terraces start to form but with diffusive edges. The terrace edges become sharp only after thermal annealing, indicating the formation of atomically flat surface ideal for layer-by-layer growth (step 2).

Figure 2a shows a Hall bar that is defined by depositing an amorphous AlN mask. There are four of such structures on every 5x5 $mm^2$ area of the substrate. After LAO deposition, the Hall bar region (which is not covered by AlN) becomes conducting while the rest remains insulating. Figure 2b shows a schematic of the electrical connections for the Hall effect measurement including a voltage gatefor applying electric field.

The thin film growth is monitored by an in-situ RHEED. Figure 3 shows a typical RHEED pattern at room temperature (a) and at the deposition temperature before (b) and after (c) the deposition.

Following the above protocol, we have successfully fabricated LAO/STO interfaces with a LAO thickness varying from 3.5 to 4 uc. The differences in morphology between 3 and 3.5 uc LAO grown on STO are shown in Figure 4a and 4b and the RHEED oscillations during the LAO growth for 3.6 and 3.8 uc are shown in Figure 4c and 4d, respectively.

The resistance data of different samples with a LAO thickness increased in a sub-unit-cell step between 3 and 4 uc provide us a more precise critical thickness (3.65 uc) of the LAO/STO

insulator-to-metallic transition. Furthermore, the carrier density and mobility can be extracted from the field effect data, through equations (1) and (2), for different LAO thicknesses at different gate voltages, providing us the density dependence of the carrier mobility. As depicted in Figure 5, this dependence suggests that the carrier-carrier scattering plays an important role at the LAO/STO interface.

**Figure 1. Evolution of the substrate surface morphology during BHF treatment.** AFM images of (a) an as-received, (b) a BHF treated, and (c) a BHF treated and thermally annealed STO substrate. It shows selective etching and recrystallization processes makes atomically flat surfaces.

**Figure 2. Hall bar and Hall effect measurement.** (a) Optical microscopy images of the Hall bar defined by a lithographically patterned amorphous AlN layer. The "6" shape mark is at the back of the substrate. (b) The wiring layout for the Hall and electric field effect.

**Figure 3. Evolution of RHEED patterns during the LAO growth:** (a) before deposition at room temperature, (b) before deposition at 900 °C and (c) after deposition at 900 °C, respectively. The red oval enclosed spot is the spot which intensity is monitored.

**Figure 4: Sub-unit-cell control of the LAO growth on a STO substrate.** AFM images of a (a) 3 uc and (b) 3.5 uc LAO sample obtained by scanning the sample surface over the Hall bar area. The 3 uc sample shows atomically smooth terraces, while the 3.5 uc sample shows island formations on the terraces. (c) and (d) show RHEED oscillations during the growth of 3.6 and 3.8 uc of LAO, respectively.

**Figure 5: Density dependence of the carrier mobility at 2 K for samples with different LAO thicknesses.**

**DISCUSSION:**

The sample fabrication protocol described above allows sub-unit-cell control of LAO growth on a STO substrate in LAO/STO heterojunctions as shown in Figure 4. The growth control relies on the *in-situ* RHEED monitoring in a PLD system, and this protocol might be applicable to other material systems which exhibit a layer-by-layer growth mode. For materials having other growth modes, such as island growth, there is no periodic oscillation of the RHEED intensity expected, and thus sub-unit-cell control cannot be achieved with this method.

The introduction of AlN as a sacrificial mask is a reliable and robust technique to create sub-micron patterns on oxide heterostructures. This technique is much simpler and avoids the possibility of a defect formation under the etched oxide layer, which is commonly observed during ion-milling and may introduce a redundant conducting surface. The approach here would be applicable to create sub-micron structures in other (oxide) interfaces and heterojunctions.

The electric field effect is a powerful technique to continuously tune carrier density (see Figure

5 where carrier density is tuned from 1 x $10^{13}$ – 5 x $10^{13}$ $cm^{-2}$) of a material system without changing its structural and chemical composition. In this study, electric field effects using a back gate configuration with STO as the dielectric is utilized. Even though a large gate voltage is typically needed, the main advantage of such a configuration is that the STO substrate exhibits a large dielectric constant, in particular at low temperatures below 60 K, allowing a carrier modulation of up to 5 × $10^{14}$ $cm^{-2}$**.** A larger carrier modulation of up to 1 × $10^{15}$ $cm^{-2}$ with a low gate voltage (of a few volts) can now be achieved by utilizing ionic liquid in an electronic double layer transistor (EDLT) configuration, a technique that is becoming popular nowadays.

In this protocol, the critical step is the photolithography process. The UV exposure time and development time of the photoresist should be carefully controlled. If it is underdeveloped, there is some photoresist residual left on the substrate. As a result, acetone will lift off a larger area than the desired Hall bar pattern, causing the conducting path to not be defined properly. The poorly defined conducting channels affect the transport measurement results. To solve this problem, exposure and develop time have to be optimized experimentally.

In conclusion, the experimental protocol discussed in this paper allows a sub-unit-cell control of the LAO layer and sub-micron patterning in the LAO/STO interfaces. The electrical characterization of samples prepared by following this protocol provide a better understanding on the scattering mechanism of the 2DEG in the vicinity of the metal-to-insulator transition. This protocol should be applicable to other (oxide) interface systems.

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**DISCLOSURES:**

The authors have nothing to disclose.

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Figure 1
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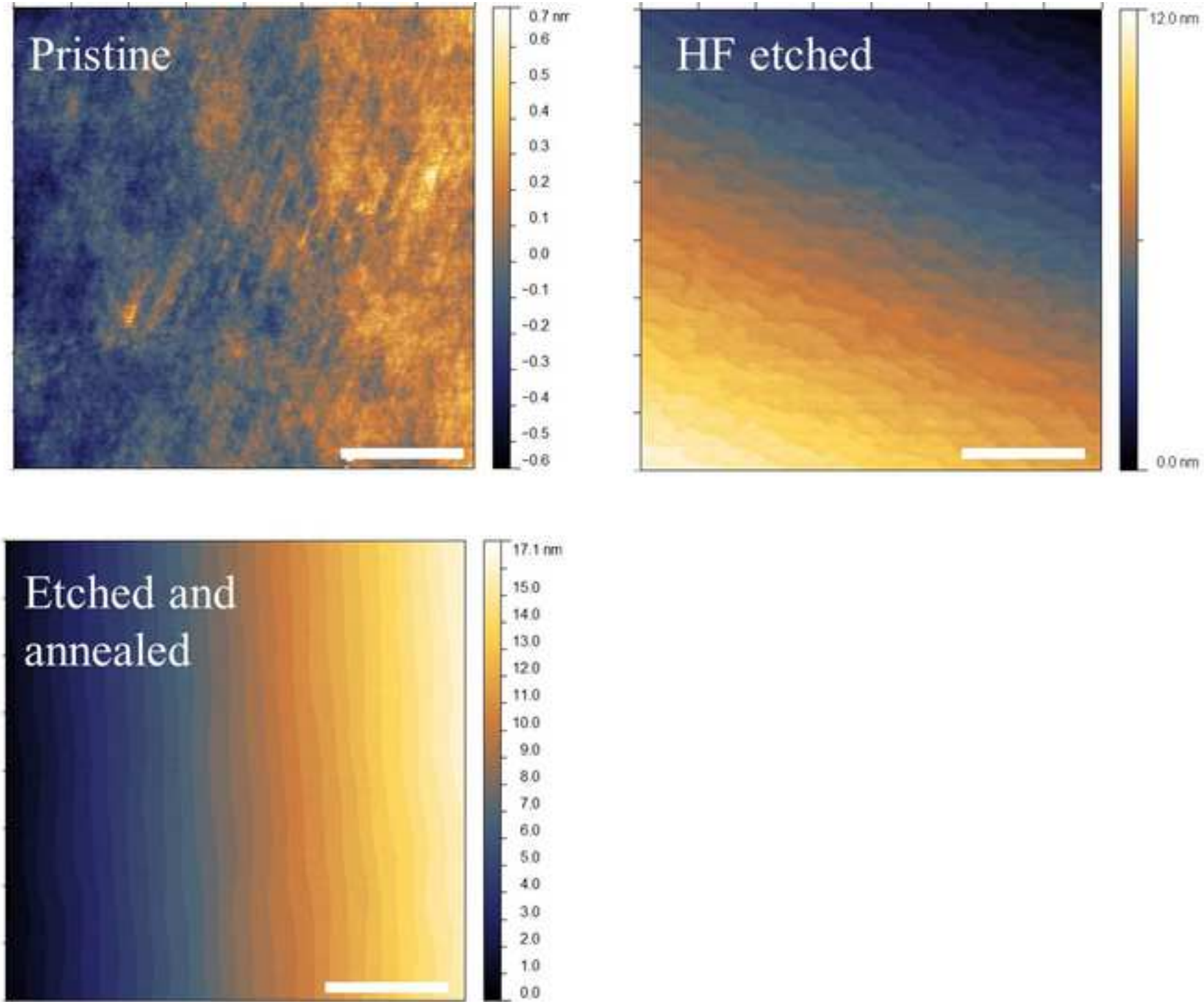

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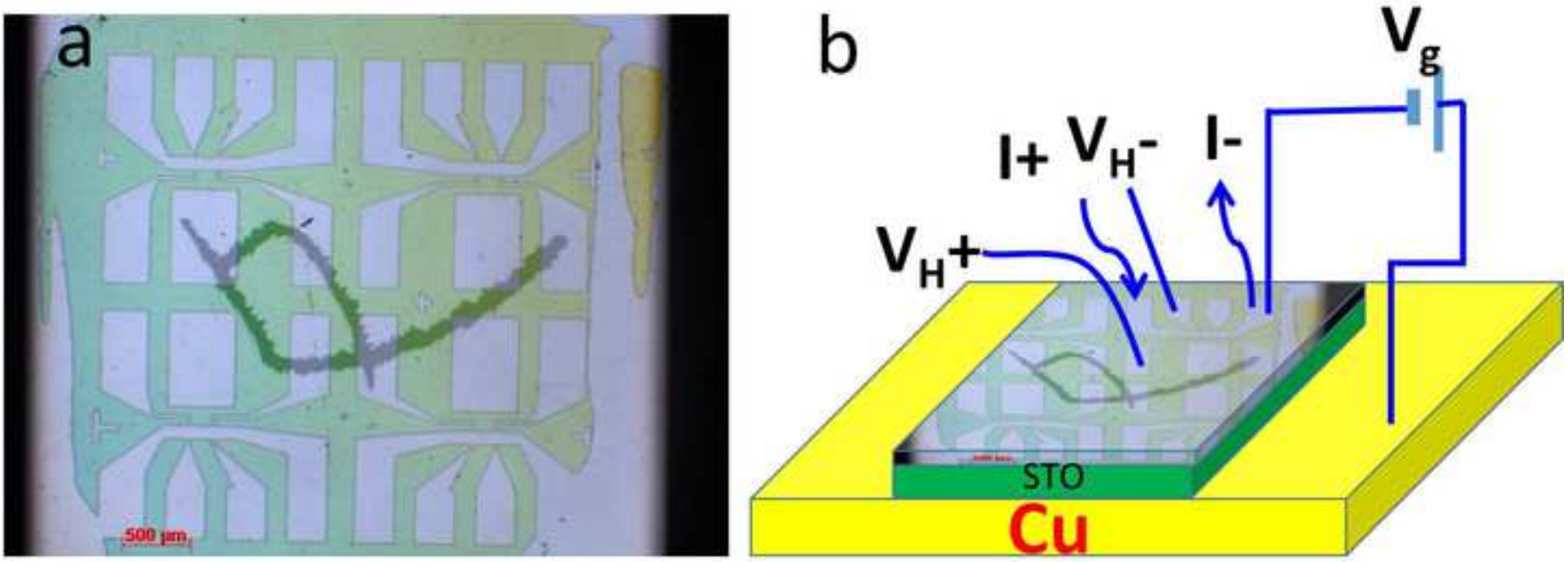

Figure 3
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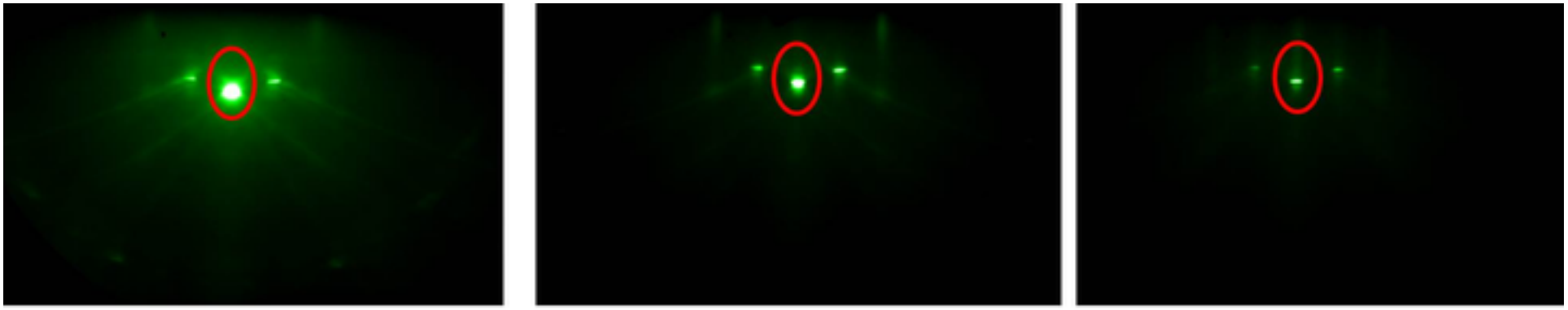

Figure 4
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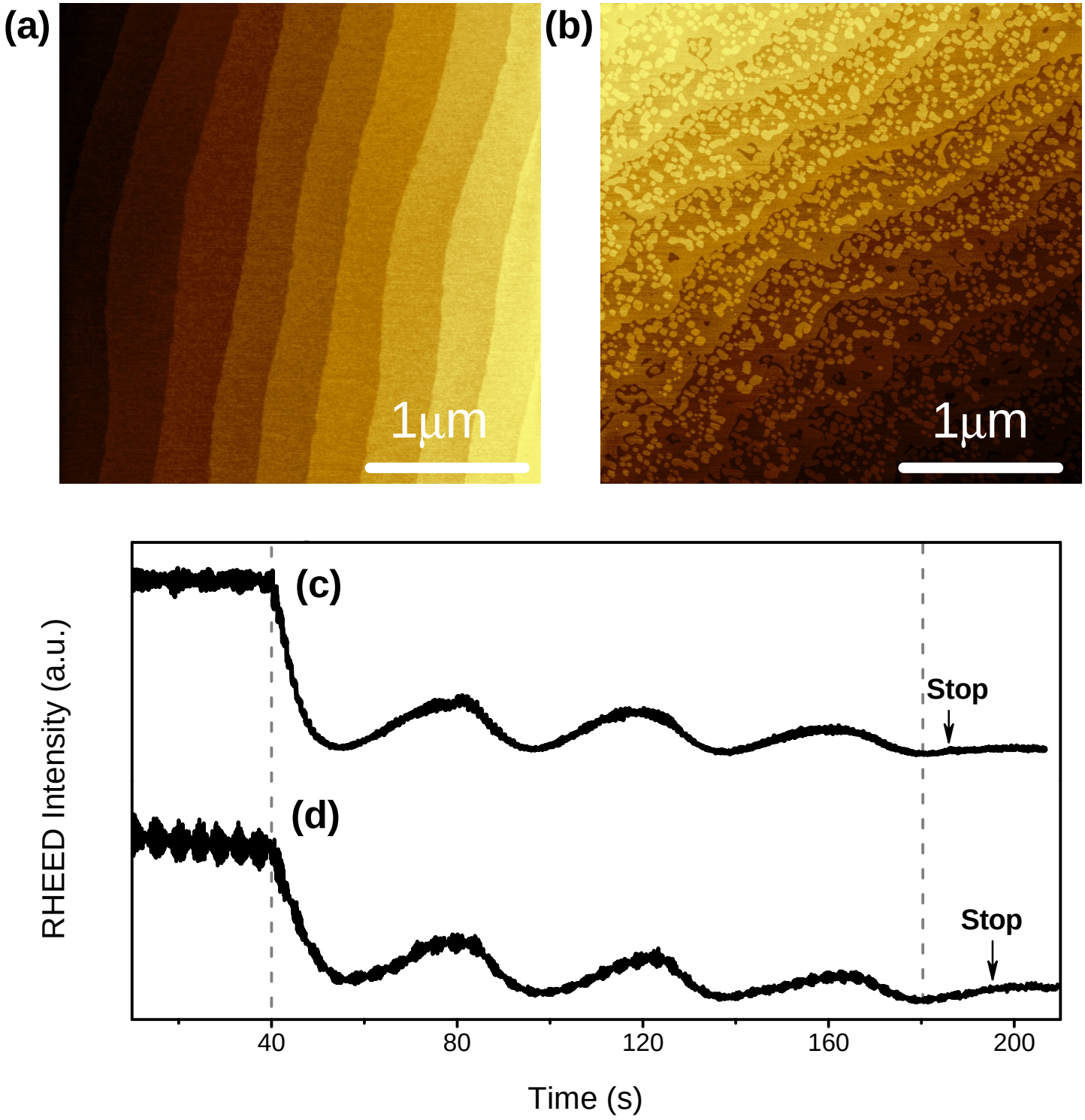

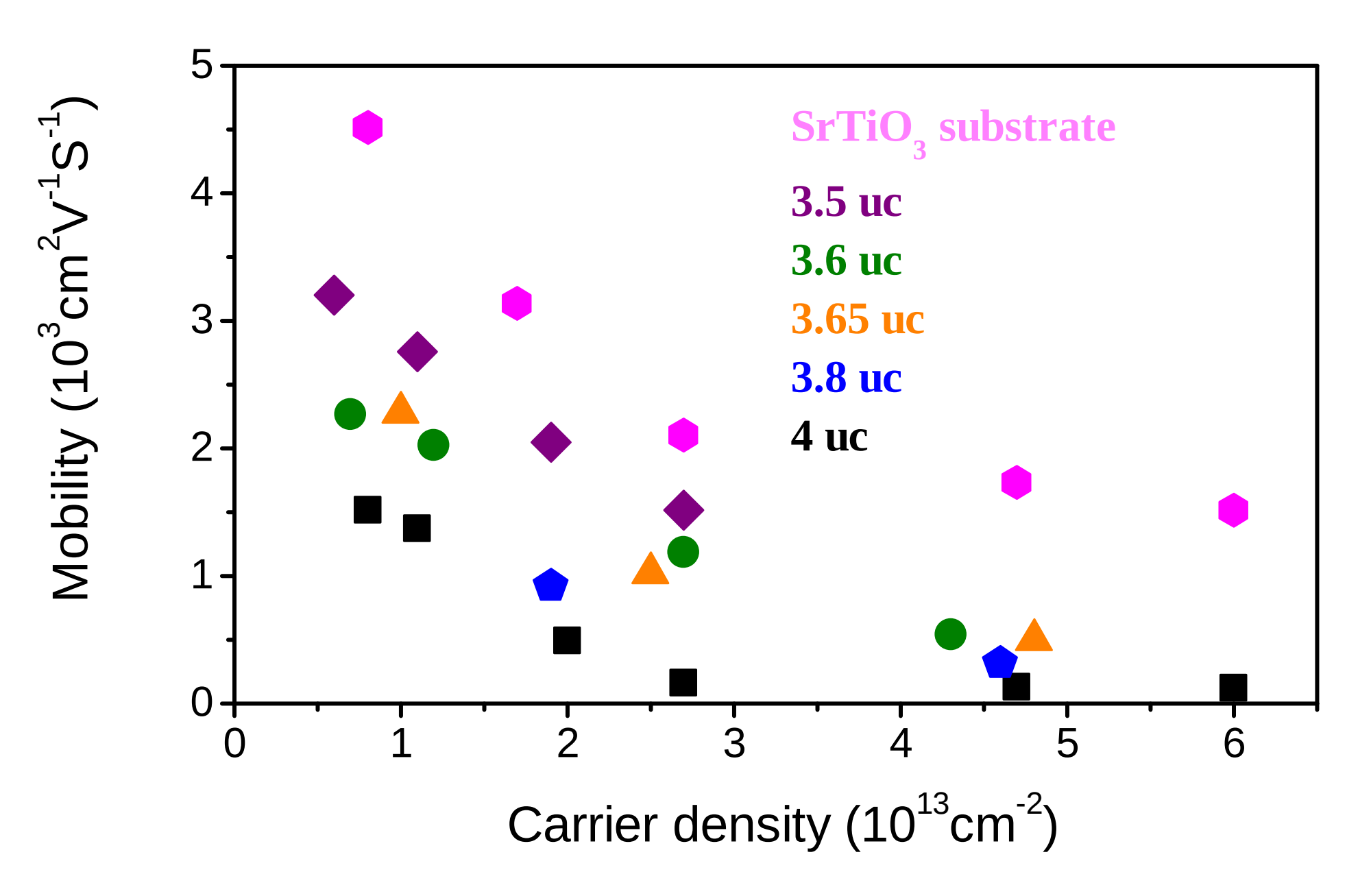
Mobility ($10^3 cm^2 V^{-1} S^{-1}$)
Carrier density ($10^{13} cm^{-2}$)
0
1
2
3
4
5
6
$SrTiO_3$ substrate
3.5 uc
3.6 uc
3.65 uc
3.8 uc
4 uc

Excel Spreadsheet- Table of Materials/Equipment

Click here to download Excel Spreadsheet- Table of Materials/Equipment: JoVE_Materials.xls

| Name of Material/ Equipment | Company | Catalog Number |
|---|---|---|
| (001) STO substrates | Crystec | NA |
| LAO single crystal target | Crystec | NA |
| Buffered HF solution | MicroChemicals | NA |
| AlN sintered target | Kurt J. Lesker | EJTALNX282A4 |
| Photoresist | AZ Electronic Materials (Germany) GmbH | AZ5214 |
| | | |
| PLD system | Neocera | NA |
| PPMS system | Quantum Design | NA |
| Kethely 2400 sourcemeter | Kethely | NA |
| Photolithography mask aligner | | NA |
| Wire bonder | West Bond | 7476D-79 |

**Comments/Description**

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Title of Article: Controlled fractional unit cell growth of $LaAlO_3/SrTiO_3$ interface near the critical thickness and its scattering properties

Author(s): Li, Changjian; Zeng, Shengwei; Liu, Weiming; Ariando; Venkatesan, T.

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Institution: National University of Singapore

Article Title: Controlled fractional unit cell growth of $LaAlO_3/SrTiO_3$ Interfaces near the critical thickness and its scattering properties

Signature: Li Changjian Date: 12 March 2015

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**Response to Editorial comments:**

The manuscript has been modified by the Science Editor to comply with the JoVE formatting standard. Please maintain the current formatting throughout the manuscript. The updated manuscript (53413_R4_051215.docx) is located in your Editorial Manager account. In the revised PDF submission, there is a hyperlink for downloading the .docx file. Please download the .docx file and use this updated version for any future revisions.

Changes made by the Science Editor:

1. There have been edits made to the manuscript.

Changes to be made by the Author(s):

1. Please revise the highlighting of the protocol to be 2.75 pages or less.

2. Grammar:

Please correct the grammar in the last sentence of the long abstract.

4.2 – Should be “using the same process as 3.2” rather than “using the same step.” Also, do you mean 3.1 instead of 3.2?

First sentence, third paragraph of the discussion – “is a powerful” what? A word is missing.

3. Formatting:

Long abstract – Please define CMOS

1.5.2 – Please define rms.

For above comments, all changes have been made accordingly in the modified manuscript.

4. All figures should have a title and short description.

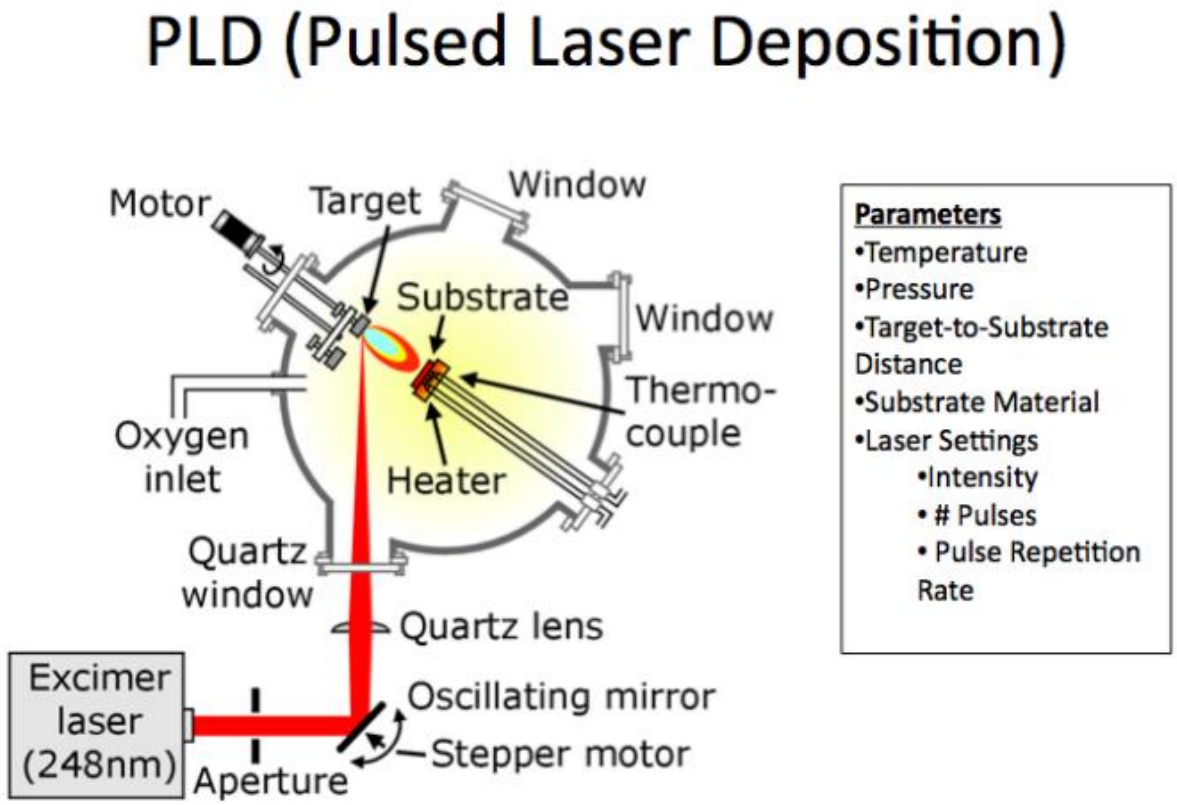
Figure caption serves the purpose, so no changes are made.

5. Additional detail is required:

3.1 – How is the AlN target secured to the holder? What is the AlN target? Is this different from the STO and the substrate holder?

3.5 – Is the substrate ablated by the laser? This is not clear.

To answer these two questions, I included a figure for PLD setup in figure R1. Hope this clears the doubts. The basic process is that: laser ablates the target (AlN and LAO in our case) surface, causing a small amount of target material to vaporize and expand towards to the substrate surface. Then the vaporized materials adsorb on substrate surface and grow into thin films. Whether thin film is crystalline or amorphous depends on the temperature of the substrate. In our study, AlN thin film is amorphous because it is deposited at room temperature while LAO is crystalline because of high temperature deposition.




(Adapted from http://www.coroflot.com/GideonGrossman/Thin-film-metal-oxide-solar-cells)

Figure R1 A schematic setup for PLD deposition.

4.5 – Which patter in figure 3 is the good one?

Figure 3 shows good RHEED patterns for different thin film growth states.

6.6 – How is this observed?

It is read through the multimeter which has the same connection for application of the gate bias.

6. References: Please abbreviate all journal titles.

Changes have been included in the modified manuscript.

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