Creation of a two-dimensional electron gas and conductivity switching of nanowires at the LaAlO3/SrTiO3 interface grown by 90° off-axis sputtering


Citation: Applied Physics Letters 103, 071604 (2013); doi: 10.1063/1.4817921
View online: http://dx.doi.org/10.1063/1.4817921
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/103/7?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Tunable bilayer two-dimensional electron gas in LaAlO3/SrTiO3 superlattices

Modulation of conductance and superconductivity by top-gating in LaAlO3/SrTiO3 2-dimensional electron systems

Non-conducting interfaces of LaAlO3/SrTiO3 produced in sputter deposition: The role of stoichiometry
Appl. Phys. Lett. 102, 121601 (2013); 10.1063/1.4798828

Gigahertz-frequency operation of a LaAlO3/SrTiO3-based nanotransistor
Appl. Phys. Lett. 102, 103113 (2013); 10.1063/1.4795725

Electron transport at interface of LaAlO3 and SrTiO3 band insulators
J. Appl. Phys. 113, 093709 (2013); 10.1063/1.4794057
Creation of a two-dimensional electron gas and conductivity switching of nanowires at the LaAlO$_3$/SrTiO$_3$ interface grown by 90° off-axis sputtering


Department of Materials Science and Engineering, University of Wisconsin, Madison, Wisconsin 53706, USA
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA
Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, Pennsylvania 15260, USA

(Received 6 October 2012; accepted 9 July 2013; published online 12 August 2013)

Two-dimensional electron gas (2DEG) formed at the interface between two oxide band-insulators LaAlO$_3$ and SrTiO$_3$ raises the possibility to develop oxide nanoelectronics. Here, we report the creation of a 2DEG at the LaAlO$_3$/SrTiO$_3$ heterointerfaces grown by 90° off-axis sputtering which allows uniform films over a large area. The electrical transport properties of the LaAlO$_3$/SrTiO$_3$ heterointerface are similar to those grown by pulsed laser deposition. We also demonstrate room-temperature conductive probe-based switching of quasi-one-dimensional structures. This work demonstrates that a scalable growth process can be used to create the two-dimensional electron gas system at oxide heterointerfaces.

Oxide heterostructures have vast potential to revolutionize electronic devices due to their high degree of tunability, functionality, and stability in extreme environments. In recent years a conducting state was reported at the interface between the two band insulators, LaAlO$_3$ (LAO), and SrTiO$_3$ (STO). This discovery of a two-dimensional electron gas (2DEG) has brought about innovative applications in the area of nanoscale oxide devices. Currently, pulsed laser deposition (PLD) is the dominant deposition technique used for creating oxide interfacial 2DEG heterostructures, and only more recently has molecular beam epitaxy (MBE) been used to create a 2DEG. In order for these devices to be broadly applicable they need to be integrated with current fabrication technology—most notably there is a need to deposit these thin films uniformly on large wafers.

90° off-axis sputtering is a scalable process that has been shown to create smooth, epitaxial films that are uniform over a large area. This is essential for integration with the Si industry. In addition, using an alternative growth technique such as sputtering can bring further insights to 2DEG research. While several groups have grown LAO/STO heterostructures by sputter deposition, none have reported a conducting interfacial 2DEG. This task requires finely tuned growth parameters that avoid having the bulk substrate conducting and the entire sample insulating while maintaining a 2DEG at the interface. In this letter, we demonstrate the creation of a conducting interface in structures grown using 90° off-axis sputtering. The heterostructures also exhibit room-temperature conductive-atomic force microscopy (c-AFM) switching.

In order to create an oxide interfacial 2DEG, we use a different regime of growth conditions, deviating from the previously reported 3:4 O$_2$:Ar ratio and high pressure O$_2$ sputter environments. In the case of PLD, it has been shown that too high (greater than 7.5 mTorr) oxygen pressure can degrade LAO structural quality and inhibit 2DEG formation. Even though oxygen vacancies are known to be removed during post-annealing after LAO growth, earlier works have shown that low oxygen partial pressure can play a major role in the creation of a 2DEG. Oxygen partial pressures used in PLD can be challenging in sputtering due to typically higher Ar gas pressure (i.e., 200 mTorr) used for 90° off-axis sputtering. There is a 3–5 order of magnitude difference in partial pressures between Ar and O$_2$, and pressure in this regime cannot be controlled accurately.

We overcome this difficulty by sputtering in pure Ar and relying on the sputtered single crystal LAO target to sustain a large enough background pressure of O$_2$ and atomic oxygen to create an environment similar to PLD. During sputtering of an oxide target material, a relatively large amount of O$_2$ and more importantly atomic oxygen is produced. The high activity of atomic oxygen compared with molecular oxygen significantly increases the effective P$_{O_2}$ during sputter growth compared with PLD. This mechanism creates an environment that can produce high quality 2DEGs during sputtering while only inputting Ar gas.

Our growth system consists of a 2-in. RF magnetron sputter source in a 90° off-axis geometry with reference to the sample heater. All films discussed here were grown on TiO$_2$-terminated (001) STO substrates. A LAO/STO control sample was grown by PLD using conditions outlined in Park et al. The sputtered samples were grown from a 9-in. single crystal LAO target mounted on a US Gun II sputter source at an RF power of 50 W. The sample temperature was 780°C during growth, consistent with previously reported works. A 200 mTorr partial pressure of Ar was used, with a minimum pre-sputter time of 15 min in order to stabilize a background partial pressure of O$_2$ (and atomic oxygen) produced from the target to be similar to P$_{O_2}$ seen in previous PLD work. This is a vital step in the growth process as films grown without this extended pre-sputtering were found to be poorly conducting.
insulating. It should also be noted that samples grown at higher P_{O2}, for example, 3:4 of O2:Ar, produce insulating films as well. We subsequently annealed the sample for 1 h in 300 Torr O2 at 600°C to ensure that the bulk substrate was not conducting. While MBE and PLD take advantage of in situ Reflection High-Energy Electron Diffraction (RHEED), which aids in the ability to have unit-cell control over the LAO growth, sputtering has no comparable in situ monitoring. Instead we have utilized x-ray reflectivity as a thickness calibration; this is essential since the thickness of LAO directly modifies the electronic characteristics of the LAO/STO interface. At the deposition conditions described above, we are able to grow LAO films at a rate of 1.65 Å/min as determined by fitting x-ray reflectivity data as shown in Figure 1(a). This data also indicate that the film is flat with small roughness.

Further x-ray analysis shown in Figures 1(b)–1(d) confirms the high-quality epitaxial growth of the LAO thin films. These high-resolution x-ray diffraction results from a 40 nm sample (Figures 1(b) and 1(c)) show an out-of-plane lattice parameter of 3.74 Å (Figure 1(b)) and in plane lattice parameter of 3.89 Å. This is close to the 3.905 Å STO in-plane lattice constant, leading to a 2.3% tensile strain in the film, referenced to a bulk LAO lattice constant of 3.790 Å. This indicates that even at 40 nm the film is almost fully strained. The phi scan in Figure 1(c) shows 4-fold symmetry of the LAO film with an in-plane (101) FWHM of 0.26° indicating a very small in-plane misalignment of the film while the in-plane FWHM of the substrate is 0.01°. As shown in Figure 1(d), a reciprocal space mapping (RSM) around the (103) Bragg peak of a STO substrate shows that the LAO film is fully coherent and of single phase. The elongation of the (10L) film peak arises from the ultra-thin thickness of only 10 unit cells of LAO. The out-of-plane lattice constant obtained from the peak position of the LAO film is 3.72 Å, which is in good agreement with that measured from the 0-2θ scan on the thicker film. The sputtered LAO film is atomically flat with an average roughness of ~0.14 nm, as shown in the atomic force microscopy (AFM) images of Figure 2. The clear step and terrace structures observed in Figure 2 arise from those of the underlying STO substrate.

A study of the carrier concentration (n_s) as a function of thickness was carried out to show the existence of a critical thickness. Samples were wire bonded with Al wires in the four-point van der Pauw geometry, and measured Hall coefficients were found to be linear as a function of applied magnetic field. Figure 3(a) shows a clear transition between a...
conducting state and an insulating state at 4 unit cells, consistent with findings on PLD-grown films. There can also be seen a negligible thickness dependence for thicknesses greater than 4 unit cells. This is a clear indication that these LAO/STO samples produced by sputtering exhibit a 2DEG behavior as explained by electronic reconstruction.

Transport measurements were carried out as a function of temperature on both sputtering and PLD samples with similar growth conditions for comparison with LAO thicknesses of 10 unit cells in the sputtered sample. The carrier density in the sputtered sample was found to have a somewhat weaker dependence on temperature than the PLD grown sample (Figure 3(b)). The temperature dependence of the Hall mobility ($\mu_H$) is similar above 75 K, where both obey a power law behavior of $T^{-\alpha}$, with $\alpha = 2.3$ and 2.4 for the sputtered and PLD grown sample, respectively (Figure 3(c)), consistent with reported behaviors. At low temperatures the mobility in the sputtered sample saturates at lower values compared with the PLD grown sample, suggesting increased scattering at these growth conditions. The increased low-temperature sheet resistance ($R_S$) is consistent with the reduced mobility values since the variation in carrier density is small in this range; in the range above 50 K both samples display similar metallic behavior (Figure 3(d)).

We also demonstrate room-temperature c-AFM creation of 2DEG nanostructures formed at atomically sharp LAO/STO heterointerfaces grown by 90° off-axis sputtering. The ability to write and erase nanostructures in these samples has been observed at a range of thicknesses between 3 and 3.7 unit cells. After 90° off-axis sputtering growth, electrically conducting contacts to the interface are defined by optical lithography. Then the sample is prepared by initially milling 25 nm deep trenches through the LAO layer via an Ar-ion mill and then filling them with 4 nm of Ti followed by 25 nm of Au to form bilayer electrodes via sputtering. Within the 30 μm × 30 μm “canvas” defined by the electrical contacts, nanostructures are written and erased at the interface using c-AFM lithography at room temperature. The conductance between two electrical contacts is continuously monitored by a lock-in amplifier (Figures 4(a) and 4(b)). Figure 4 shows a typical c-AFM writing and erasing process in a 3.4 unit cell LAO/STO sample with an initially insulating interface. To begin with, two rectangular pads are “written” (that is, raster scanning by a $V_{\text{write}} = +10$ V biased AFM tip) for better contacting to the two Au electrodes, then a conductive nanowire is created by the +10 V biased AFM tip scanning from one electrode to another at 300 nm/s speed (Figure 4(a)). When the tip reaches the other electrode, a pronounced and abrupt conductance jump is observed (Figure 4(c)). The observed maximum conductance change and non-exponential decay in atmosphere conditions are comparable with the 3.4 unit cell LAO/STO samples grown by PLD. After writing the nanowire, the AFM tip is repositioned and biased at $V_{\text{erase}} = -10$ V, then moves perpendicularly across the nanowire at 10 nm/s speed (Figure 4(b)). The conductance decreased abruptly to zero when the tip reaches the nanowire (Figure 4(d)). The nanowire width can be quantified by fitting the conductance drop curve with the function $G(x) = G_0 \cdot G_1 \cdot \tanh(x/h)$, the red curve in Figure 4(d). From the best fitting parameters we get a nanowire width of 9.2 nm. The ability to create conductive nanostructures is important for technological applications, but it also serves as a sensitive probe of the nanoscale uniformity of the 2DEG. A single insulating patch along the nanowire is sufficient to prevent conductivity. Here, the properties are comparable to PLD-grown heterostructures.

Sputtering is a thin film growth technique capable of the uniform large area deposition required for scaling. In this
work we have demonstrated that sputtering can be used to grow high-quality epitaxial LaAlO$_3$ films with excellent surface quality on TiO$_2$-terminated SrTiO$_3$ substrates. Growth conditions were chosen to mimic those used by growth processes known to form a 2DEG at the interface, and this resulted in the creation of a LaAlO$_3$/SrTiO$_3$ two-dimensional electron gas made by sputtering. We showed that these samples have comparable electronic transport properties to heterostructures grown with pulsed laser deposition. We have also demonstrated room-temperature conductive-AFM switching of these 2DEG nanostructures showing the capability to make interesting oxide devices on these films. In summary, we have created another avenue of exploration for the study of the 2DEG at the LaAlO$_3$/SrTiO$_3$ interface by developing a growth process using a sputter deposition method.

The authors wish to acknowledge financial support from the National Science Foundation (NSF) under Grant Nos. DMR-1234096 (CBE), DMR-0906443 (CBE), DMR-1104191 (JL), DMR-1124131 (CBE, JL) and from AFOSR under Grant Nos. FA9550-12-1-0342 (CBE), FA9550-10-1-1104191 (JL), DMR-1234096 (CBE), DMR-0906443 (CBE), DMR-1124131 (CBE, JL), and from AFOSR Grant Nos. FA9550-12-1-0342 (CBE), FA9550-10-1-1104191 (JL), DMR-1234096 (CBE), DMR-0906443 (CBE), DMR-1124131 (CBE, JL), FA9550-12-1-0268 (JL).

1J. Mannhart and D. G. Schlom, Science 327(5973), 1607 (2010).