Nanoscale patterning of electronic devices at the amorphous LaAlO3/SrTiO3 oxide interface using an electron sensitive polymer mask

Bjorlig, Anders V.; von Soosten, Merlin; Erlandsen, Ricci; Dahm, Rasmus Tindal; Zhang, Yu; Gan, Yulin; Chen, Yunzhong; Pryds, Nini; Jespersen, Thomas S.

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Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, Universitetsparken 5, 2100 Copenhagen, Denmark
Department of Energy Conversion and Storage, Technical University of Denmark, Risø Campus, 4000 Roskilde, Denmark

A simple approach is presented for designing complex oxide mesoscopic electronic devices based on the conducting interfaces of room temperature grown LaAlO$_3$/SrTiO$_3$ heterostructures. The technique is based entirely on methods known from conventional semiconductor processing technology, and we demonstrate a lateral resolution of ~100 nm. We study the low temperature transport properties of nanoscale wires and demonstrate the feasibility of the technique for defining in-plane gates allowing local control of the electrostatic environment in mesoscopic devices. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).

Since the discovery of a two-dimensional conducting electron system (2DES) at the interface between TiO$_2$ terminated strontium titanate (STO) and lanthanum aluminate (LAO), the efforts towards exploring, understanding, and utilizing the electronic properties of oxide interfaces have attracted significant attention from all branches of physics and materials science. One example is the drive towards the integration of oxide interfaces in mesoscopic devices to complement the semiconductor heterostructures and bottom-up grown nanostructures which have been the material platform for two decades of research in quantum transport. The oxide electron systems share the properties of low dimensionality and electrostatic gatability with the conventional semiconductor systems but in addition display a wide range of phenomena such as gate-tunable superconductivity, ferroelectricity, magnetism, and spin-orbit coupling caused by strong electron-electron and electron-lattice interactions in regimes not accessible in semiconductors. However, the oxide 2DES is highly sensitive to the quality of the epitaxial interfaces and subsequent surface processing, and a range of specialized fabrication techniques have therefore been developed for realizing patterned oxide devices while protecting interfaces and limiting post processing. Such techniques range from patterned growth of the oxide top-layer by hard-mask techniques, ion irradiation, thickness variations of the top layer, or the creation of conducting nanostructures using scanning probe lithography. Such approaches are more involved than conventional semiconductor processing and often include wet etching which is difficult to control leading to limited reproducibility and resolution.

Most studies so far have considered crystalline STO-based heterostructures where the top-films are deposited at temperatures above ~600 °C. Room temperature (RT) deposition such as for the amorphous LAO/STO (a-LAO/STO) system also leads to conducting interfaces where electrons predominately originate from oxygen vacancies, and usually exhibit reduced mobilities compared to crystalline LAO/STO. The key properties of gate-tunable and gate-tunable superconductivity, however, remain intact, and moreover, the highest transition temperature reported for the STO-based 2DES was achieved in the a-LAO/STO system. Also, recently, studies were performed on the transport properties of negative-U quantum dot devices which were fabricated from a-LAO/STO heterostructures. Thus, the a-LAO/STO system provides an interesting system for mesoscopic oxide devices, and here, we demonstrate that the reduced deposition temperature allows patterning of the interface conductivity by a conventional electron-beam or optical lithography with only minor adjustments. We explore the limitations of the technique in terms of lateral resolution and the performance of nanoscale devices at low temperatures.

Our fabrication procedure is schematically illustrated in Fig. 1(a). The starting point is a (001) STO single crystal with a TiO$_2$ terminated surface achieved by first sonicating in ethanol and acetone at room temperature (RT) for 300 s, then for 20 min in deionized (DI) water at 70 °C, and 20 min in aqua regia (3:1:16 HCl:HNO$_3$:H$_2$O) at 70 °C. Following a brief 30 s cleaning in DI water, the substrates are annealed for 60 min at 1000 °C in a tube furnace under oxygen flow and cooled to RT with a ramp rate of 100 °C/h. Substrates for e-beam(optical) lithography are then spin-coated with 2% polymethyl methacrylate (PMMA) diluted in anisole(AZ1505 photo resist) and heated for 1 min at 185 °C(115 °C) on a hot plate. Additionally, the e-beam substrates are spin-coated with a layer of SX-AR-PC 5000 e-spacer, which is heated for 60 s at 115 °C, to reduce distortion during exposure. PMMA(AZ1505) was exposed using standard parameters and the patterns developed in 1:3 Methyl-Isobutyl-Ketone and Isopropanol(MF-321 developer). Before growing the top-layer, the substrates were carefully cleaned in an oxygen plasma for 120 s (calibrated to remove 8 nm of PMMA) and cleaned in Milli-Q (MQ) water for 120 s at RT. Lift-off patterning of a-LAO combined with subsequent high-resolution lift-off...
temperature annealing and deposition has previously been used for defining insulating structures in crystalline LAO/STO devices. For defining conducting structures, it is, however, crucial that the exposed surface does not contain any resist residues and previous attempts excluding the last cleaning steps have been found to lead to insulating samples. We have tested the patterning technique for both e-beam lithography and optical lithography yielding similar results; in the following, we present the results obtained using the former technique. The final PMMA resist profile is shown in Fig. 1(e), and a representative AFM image of the surface terraces in the exposed channel is shown in Fig. 1(f) after the cleaning procedure. No discernible differences to the as-treated surface are observed. A top layer of 12 nm a-LAO was grown by room temperature Pulsed-Laser-Deposition (PLD). To investigate the properties of the 2DES induced at the exposed interfaces and the spatial resolution of the lithographical patterning, standard Hall-bars (Fig. 2) were defined as well as structures allowing 4-terminal measurements of leakage currents between parallel wires with varying separations [Fig. 3(a)] and of the conductance of wires of varying widths [Fig. 3(c)].

Hall-bar samples were fabricated with a width of \( W = 30 \mu m \) and a length of \( L = 500 \mu m \), and Fig. 2(a) shows a representative sample after the growth of the top layer. Measurements were performed in a dilution refrigerator with a \( \sim 20 \) mK base temperature, and standard lock-in techniques (\( \sim 20 \) nA current excitation) were used to measure the longitudinal \( V_{xx} \) and transverse \( V_{xy} \) voltage drops as indicated in the figure. The samples were mounted in a sample carrier using conductive silver epoxy, and the overall carrier density was modulated by biasing the conducting back-plane of the sample (\( V_{BG} \)). No leakage was measured between neighboring Hall-bars on the samples. As seen in Fig. 2(b), the sheet resistance \( R_s = (W/L)V_{xx}/I \) decreases upon cooling as typically observed for metallic oxide interfaces. The dependence of \( R_{xx} \) and \( R_{xy} \) on a perpendicular magnetic field \( B \) at 20 mK is shown in Fig. 2(c). The positive magneto-resistance in \( R_{xx} \) is observed in all samples and has also been reported previously for oxide interfaces and has been attributed to contributions from more than one carrier type. The finite value for \( R_{xy} \) at \( B = 0 \) is attributed to a contribution from the longitudinal voltage drop due to an off-set of the voltage probes or a non-isotropic current path in the sample. Fitting \( R_{xx} \) and the high-field value of \( R_{xy} \) to the two-band model as shown in the figure, we find a higher density \( n_1 = 4.75 \times 10^{13} \text{cm}^{-2} \) of low mobility \( \mu_1 = 125 \text{cm}^2/\text{V s} \) and a lower density \( n_2 = 7.5 \times 10^{12} \text{cm}^{-2} \) of high mobility carriers.
Hall-bar devices were fabricated on 3 substrates (6 patterned ing reason for this variation remains unknown. In total, 18 variations when substrates exhibit superconductivity. The underly- unpatterned growths, we find significant growth-to-growth var- 

The spatial resolution of the patterning technique is cru- 

This indicates that the patterning process, in particular, the oxy- 

As is common for LAO/STO samples, we find that electro- 

Considering first the leakage between separated struc- 

Figure 3(d) shows the RT resistance of the wire devices as a function of the width. To reduce the effects of inhomoge- 

Each wire is 100 μm long and has two 45 μm long sections that are kept at a width of 5 μm joined together by a central narrow section of length L and varying width W. In
of the 2DES and summarize, the results in Fig. 3 show that within an uncertainty of 25 nm, the separation between the gates and the channel was likewise 500 nm. The conductance of the devices as a function of the voltage $V_{SG}$ applied to the side-gates and back-gate is shown in Figs. 4(b)–4(d) for a temperature of 20 mK. An interesting regime is that close to pinch-off where only a few channels contribute to the transport, phenomena such as quantized conductance are expected. For all three gates, the conductance significantly decreases with lowering gate potential as the channel is depleted of carriers and the effect of the two side gates is remarkably symmetric. The gate-efficiency, however, progressively decreases at lower gate voltage due to the field dependent dielectric constant of STO, and the side-gates could be operated without leakage only within the range of $[-0.15 \text{ V}, 1 \text{ V}]$. A similar device with lower dimensionality was fabricated after these measurements. Here, conductance was observed through a 100 nm channel with a non-leaking interval of $[-0.15 \text{ V}, 0.15 \text{ V}]$ and similar results from the transport measurements. Although the devices were operated in the regime close to pinch-off, no quantization of conductance nor signatures of conductance fluctuation were observed. This is similar to previous reports and is interpreted as a current carried by many weakly transmitting transverse modes. Systematic investigations of device geometries varying the potential steepness around the channel may shed further light on this.

In conclusion, we have presented a patterning approach for the fabrication of mesoscopic devices from STO-based heterostructures with room temperature grown top-layers. The technique utilizes only conventional polymer resists sensitive to either electron or optical exposure and allows for patterning with a resolution of at least 100 nm without affecting the quality of the 2DES. Finally, we demonstrated devices implementing local electrostatic gate-control of the carrier density in narrow constrictions. Various aspects of the patterning approach were demonstrated using room temperature grown amorphous LAO as the top-layer; however, alternative STO-based conducting room temperature grown interfaces have been reported and we expect that the techniques reported in this study may be directly applied to such systems as well. The results present a simple way to bridge the gap between oxide-based quantum device fabrication and state-of-the-art semiconductor processing.

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