Microlithography of electron gases formed at interfaces in oxide heterostructures

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Submicron wide structures of conducting quasi-two-dimensional electron gases generated at $SrTiO_3/LaAlO_3$ interfaces have successfully been patterned by modulating the thickness of the $LaAlO_3$ layers with unit cell resolution. This technique allows the authors to structure the electron gases without exposing them to the environment and without incorporating other materials at the edges. The structured electron gases have resistances of $\approx 200\ \Omega/\square$ (4.2 K) and mobilities of $\approx 700\ cm^2/V\ s$ (4.2 K), while the resistances of the areas patterned to be insulating exceed $10^{10}\ \Omega/\square$. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2354422]

As shown by Hwang and co-workers,[1,2] conducting electron gases may form at interfaces in heterostructures that consist of insulating oxides such as $SrTiO_3$ and $LaTiO_3$ or $LaAlO_3$ and $SrTiO_3$. These electron gases are confined to sheets that are only very few nanometers thick. We therefore call them quasi-two-dimensional electron gases (q2-DEGs). Their charge carriers have been found to have mobilities as high as $10^4\ cm^2/V\ s$ at 4.2 K.[1,3,4]

All studies of these electron gases have been performed with unpatterned samples, i.e., the electron gases were generated over the complete sample areas and were measured as such. However, most studies and possible applications require that the gases are lithographically patterned. Unfortunately, standard lithography techniques are not applicable to such structures. The straightforward solution (patterning by Ar-ion etching) is, for example, not useful, because the damage created by the ion beam causes the $SrTiO_3$ to become conducting. The conducting $SrTiO_3$ then shunts the electron gases. It is obviously desirable to pattern the electron gases without exposing the edges of the conducting channels to the environment or to chemicals to avoid interactions between the electron gas and the defect-rich layers that then exist at the edges.

Here we present a lithography technique that allows us to pattern the electron gases without creating unwanted shunts. It also does not open the interface during the patterning process. This technique is based on the recent discovery that for $LaAlO_3/SrTiO_3$ interfaces the conductivity of the electron gases changes in a steplike manner by more than seven orders of magnitude from insulating to metallic behavior if the $LaAlO_3$ sheets reach a critical thickness of $d_c = 4$ uc (unit cell).[5] Due to the existence of this critical thickness the electron gases present at the $LaAlO_3/SrTiO_3$ interfaces can be patterned in a straightforward way. By growing the $LaAlO_3$ layers to a thickness $d < d_c$ in parts of the sample, these parts remain insulating. Likewise, areas of the samples with $LaAlO_3$ layers of thickness $d \geq d_c$ generate mobile electron gases and therefore are conducting.

To explore this patterning technique, $LaAlO_3$ layers were grown by pulsed laser deposition on TiO_2 terminated (001) $SrTiO_3$ surfaces[6] using high oxygen pressure reflection high-energy electron diffraction (RHEED) to control the layer thickness on a unit cell level.[7] The films were deposited from stoichiometric, single crystalline targets in an oxygen atmosphere of 2×10^{-5} mbar at 770 °C, and then cooled to room temperature in 400 mbars of O_2. The 2.5 h cooldown included a 1 h oxidation step at 600 °C.

For the lithography of the electron gases we used a sequential deposition of epitaxial and amorphous $LaAlO_3$ layers (see Fig. 1). First, 2 uc of epitaxial $LaAlO_3$ were grown on $SrTiO_3$. Then lines with widths ranging from 1 to 100 μm were defined by standard optical UV lithography and patterned into four-point configurations. Smaller

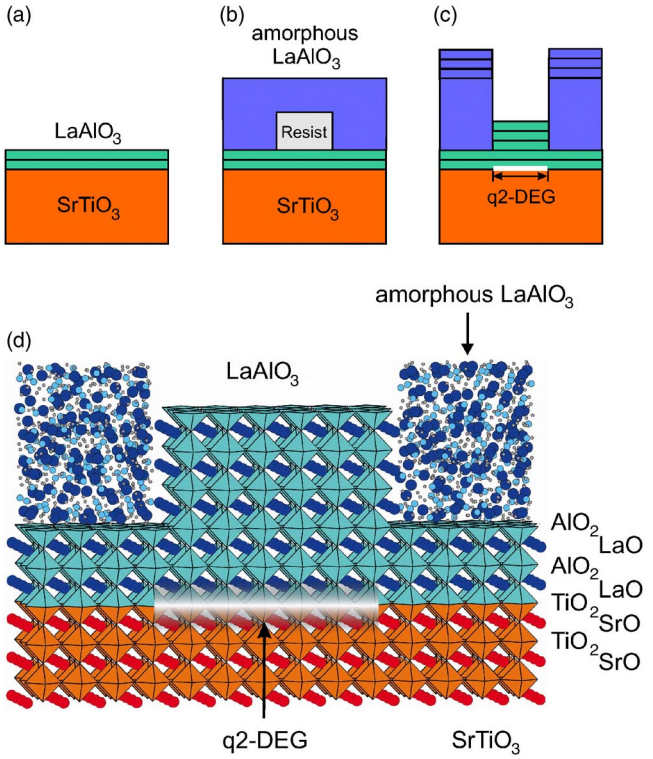

FIG. 1. (Color online) Illustration of the patterning technique. (a) On a TiO_2 terminated $SrTiO_3$ substrate 2 uc of $LaAlO_3$ are deposited epitaxially. (b) On a resist lift-off structure, 10 nm of amorphous $LaAlO_3$ are grown, and (c) then 3 uc of $LaAlO_3$ are deposited. At the $SrTiO_3/LaAlO_3$ interface the q2-DEG is formed at the areas defined in (b). (d) Sketch of a cross-sectional cut through a sample.

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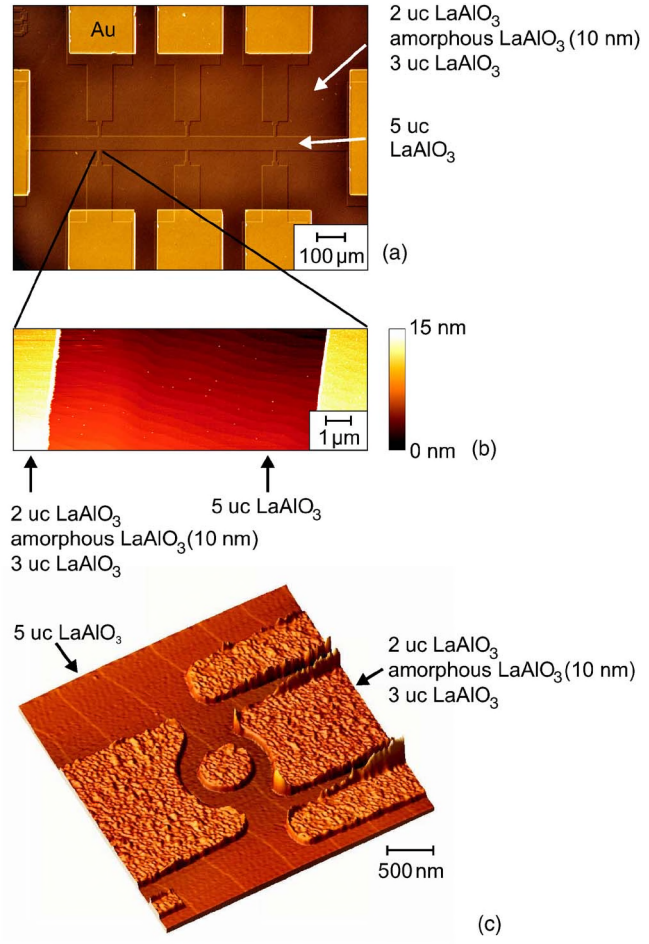

FIG. 2. (Color online) (a) Optical micrograph of a 50 μm wide track of the lithographically defined electron gas. The voltage contacts for four-point and Hall measurements are placed 300 μm apart. The height difference between the epitaxial and amorphous areas is $\approx$10 nm. (b) AFM picture of an area defined by the sequential deposition of epitaxially and amorphously grown $LaAlO_3$. (c) AFM image of a ring defining a q2-DEG with a diameter of $\approx$1.2 μm and a track width of $\approx$220 nm. Each half of the ring comprises one $LaAlO_3$ terrace.

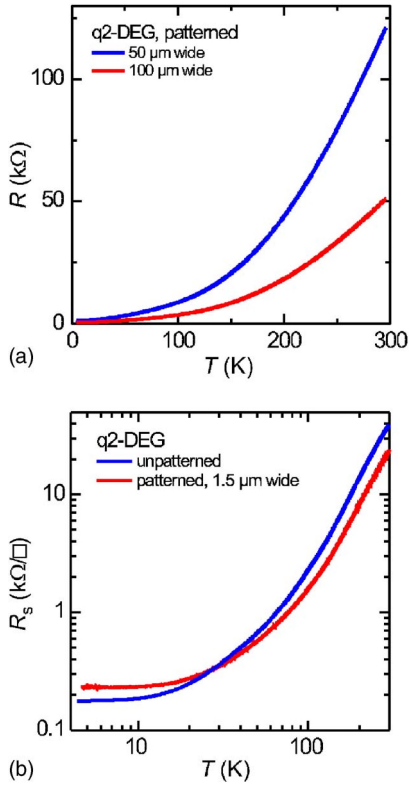

FIG. 3. (Color online) (a) Resistance of the q2-DEG measured as a function of temperature using lithographically defined tracks with widths of 50 and 100 μm and length of 300 μm (distance between the voltage probes). (b) Sheet resistance of the q2-DEG measured as a function of temperature for an unpatterned sample and for a track with a width of 1.5 μm.

structures with widths down to 200 nm were prepared using e-beam lithography. On these lift-off structures, 10 nm of amorphous $LaAlO_3$ were deposited at room temperature. After removing the resist, epitaxial $LaAlO_3$ layers were then grown in an oxygen atmosphere of 2×10^{-5} mbar at 770 °C. For the second epitaxial $LaAlO_3$ layers thicknesses between 2 and 13 uc were used. $LaAlO_3$ grows homoepitaxially on the areas that are free of amorphous $LaAlO_3$, as monitored by RHEED and by atomic force microscopy (AFM). On the amorphous $LaAlO_3$, the top $LaAlO_3$ layers grow amorphously (see also Fig. 2).

The optical microscopy image presented in Fig. 2(a) shows a structure fabricated by this method. This sample consists of conducting tracks with a width of 50 μm and a length of 1 mm, connected to six voltage pads, each 300 μm apart. The height difference between the epitaxial and amorphous $LaAlO_3$ is approximately 10 nm. The AFM image [Fig. 2(b)] characterizes the bridge edges formed at the contacts between the epitaxially and amorphously grown $LaAlO_3$. The epitaxially grown area reveals atomically flat growth terraces. The height of the steps between the terraces equals 1 uc of $LaAlO_3$. The excellent quality of this surface, which was not touched by any chemicals, allows further epitaxial growth on the patterned samples. The amorphous area is characterized by a smooth, yet irregular surface structure. The step between the amorphous and epitaxial $LaAlO_3$ is steep with a nominal width of less than 2 nm. This measured step width is limited by the sharpness of the AFM tip.[8]

We also explored a different route to lithographically pattern the electron gas. In this approach, a resist structure is directly patterned onto the TiO_2 terminated $SrTiO_3$ surface. After the deposition of amorphous $LaAlO_3$ and lift-off, 5 uc of epitaxial $LaAlO_3$ were deposited. This technique is simpler, but suffers from the drawback that the $SrTiO_3$ surface later used for the $SrTiO_3/LaAlO_3$ interface is exposed to the resist and to the solvents. The resistances of lithographically defined structures prepared such are higher than those of the previously described samples.

To contact the conducting interface which is encapsulated by the $SrTiO_3$ and $LaAlO_3$, lithographically defined contact areas were prepared by Ar-ion etching holes with a depth of $\approx$80 nm into the sample surface and by then filling these holes by an *ex situ* sputter deposition of Au. The resistances of the areas patterned to be conducting or insulating were measured with standard four-point measurements. The resistances of the insulating areas exceeded the input impedance (10^{10} Ω) of the voltmeter used (Keithley Model 2000).

Areas containing conducting electron gases are characterized by sheet conductances $\sigma\approx5\times10^{-3}$ $(\Omega/\square)^{-1}$ (4.2 K) and $\approx5\times10^{-5}$ $(\Omega/\square)^{-1}$ (300 K). As shown by Fig. 3, the resistance of the conducting electron gas scales according to the structure size. In Fig. 3(b), the sheet resistances of an unpatterned sample with 6 uc of $LaAlO_3$ and of a track with a width of 1.5 μm are shown as a function of temperature.

The electron gases of these structures could be operated with current densities as high as 3×10^6 and 2×10^4 A/cm^2 at 4.2 and 300 K, respectively. According to Hall measurements, the sign of the charge is negative with mobilities of $\approx$700 and $\approx$7 cm^2/V s at 4.2 and 300 K, respectively. These values are somewhat smaller than the values characteristic for our unpatterned samples (10^3 cm^2/V s for $n=10^{13}$ cm^2) and also smaller than the values reported in the literature.[1,3,9] We attribute these differences to the confinement of the electron gas and to the larger influence of electron scattering at the edges. Whether additional, unknown scattering centers are introduced by the structuring process is a question to be answered by further experiments.

The insulating properties of the 5 uc thick $LaAlO_3$ layers have also been characterized. Typical resistance values are $\approx$100 MΩ (4.2 and 300 K) with leakage currents $<$10 nA measured for an area of 50×100 μm^2. This corresponds to a resistivity $\rho_{LaAlO_3}\approx2\times10^8$ Ω m.

In summary, we have presented a lithography technique that allows us to pattern the q2-DEGs formed at interfaces in oxide heterostructures with submicron resolution. Neither does the technique create unwanted shunts nor does it open the interface during the patterning process. The smallest conducting structures realized were $\approx$200 nm in width.

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