Probing the electronic compressibility of $LaAlO_3$–$SrTiO_3$ interfaces by Kelvin probe microscopy

C. Richter and J. Mannhart

A two-dimensional electron liquid is formed at the *n*-type interface between $SrTiO_3$ and $LaAlO_3$. Here we report on Kelvin probe microscopy measurements of the electronic compressibility of this electron system. The electronic compressibility of the system is found to be negative for carrier densities of ≈ 10^{13}/cm^2. At even smaller densities, a metal-to-insulator transition occurs. These local measurements corroborate earlier measurements of the electronic compressibility of $LaAlO_3$–$SrTiO_3$ interfaces obtained by measuring the capacitance of macroscopic metal-$LaAlO_3$–$SrTiO_3$ capacitors.

It has been established that a two-dimensional (2D) sheet of mobile electrons is generated at the interface between the TiO_2-terminated (001) surface of $SrTiO_3$ and $LaAlO_3$ [1]. This electron system has remarkable properties that differ significantly from the properties of 2D electron gases embedded in semiconductor heterostructures. For example,the characteristic carrier density n at $LaAlO_3$–$SrTiO_3$ interfaces equals several 10^{13}/cm^2, which is well above the typical densities of 10^{11}–10^{12}/cm^2 found in semiconductor heterostructures. The charge carriers at the interface originate from an electronic reconstruction and occupy Ti 3d t_{2g} states at the interface TiO_2 layer. In the samples investigated the electron mobility is of order 1000 cm^2/Vs at 4.2 K. Moreover the system shows coexistent superconductivity and magnetism if cooled to low temperatures. Furthermore, by performing capacitance measurements on $SrTiO_3$–$LaAlO_3$–Au and $SrTiO_3$–$LaAlO_3$–$YBa_2Cu_3O_{7-x}$ capacitors, a state with negative electronic compressibility $\kappa=(n^2 d\mu/dn)^{-1}$ was identified. Here μ is the electrochemical potential [2].

In a dilute electron gas, a negative electronic compressibility results from the dominance of exchange and correlations terms, which apparently explain the negative compressibility in some semiconductor heterostructures. The origin of the observed negative compressibility of the less diluted electron liquid at $LaAlO_3$–$SrTiO_3$ interfaces has not been identified completely. The negative electronic compressibility of the $LaAlO_3$–$SrTiO_3$ interface electron system was found to exceed the negative compressibility of 2D electron gases in Si heterostructures by a factor of at least ten. It is also much larger than the negative compressibility recently reported in carbon nanotubes and GaAs structures. Although all studies of the negative electronic compressibility at $LaAlO_3$–$SrTiO_3$ interfaces were done with samples in which the $LaAlO_3$ layers were ten or twelve-unit-cells-thick to prevent tunneling currents which are unfavorable in capacitance measurements, the negative electronic compressibility has been predicted to occur also in samples with $LaAlO_3$ films as thin as four monolayers, the thinnest films to generate a conducting $LaAlO_3$–$SrTiO_3$ interface. Because tunneling and leakage currents through the $LaAlO_3$ layer undermine the accuracy of measurements of the electronic compressibility based on planar capacitors, we have explored such samples by local measurements of the electronic compressibility. For this we used Kelvin probe microscopy.

We fabricated $LaAlO_3$–$SrTiO_3$ heterostructures comprising four-unit-cells-thick (1.6 nm) epitaxial $LaAlO_3$ films. The $LaAlO_3$ films were grown by pulsed laser deposition on the TiO_2-terminated (001) surface of $SrTiO_3$ crystals. The deposition was performed at a substrate temperature of 780 °C in an oxygen background pressure of ≈ 1×10^{-4} mbar, and was monitored by reflection high-energy electron diffraction. After cooling the samples in 400 mbar O_2 , an aluminum shadow mask with a rectangular hole was attached to the samples' surfaces. Using this mask, Ar ion etching was employed to etch holes into the $SrTiO_3$ substrates, which were then filled with electron-beam-evaporated Ti to contact the interface electron liquid. After transporting the samples to the preparation chamber of the scanning probe microscope (SPM) they were heated radiatively for > 40 minutes to ≈170 °C to clean their surfaces. The SPM, which operates in ultrahigh vacuum at 4.7 K, utilizes a cantilever based on a quartz tuning fork (qPlus-sensor) with a spring constant of ≈ 1 800 N/m. An iridium spall treated *in situ* by field emission was used as a tip. The experimental setup is sketched in Fig. 1. The standard step-and-terrace structure of the $LaAlO_3$–$SrTiO_3$ heterostructures resulting from a slight vicinal cut of the $SrTiO_3$ substrates (≈0.15°) is readily imaged by scanning force microscopy (Fig. 2). Whereas on more standard samples excellent resolution was easily achieved with the SPM it was not possible to obtain atomic resolution on the $LaAlO_3$–$SrTiO_3$ heterostructures.

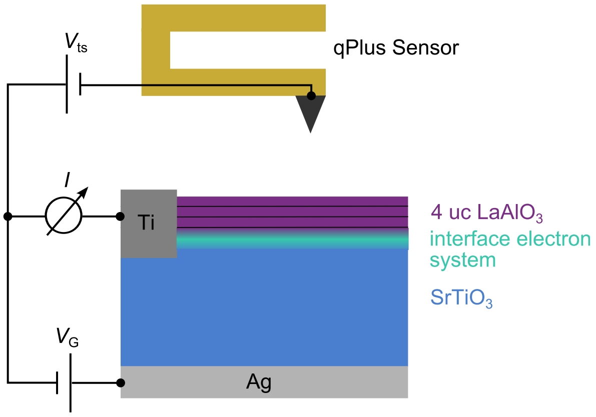

Figure 1: Illustration of the experimental configuration. The force sensor consists of a quartz tuning fork.The force between tip and sample is monitored as a function of V_{ts}, the voltage applied between tip and sample. Through the back-gate voltage V_G between the interface and the back of the $SrTiO_3$ substrate the carrier density at the interface is tuned. A negative V_G causes the n-type interface electron system to be depleted. The current I flowing into the interface and the force between tip and sample are recorded simultaneously (from [3]).

To assess the electronic compressibility of the interface we measured the force F between the tip and the sample as function of tip-sample distance, tip-sample voltage V_{ts} and back gate voltage V_G. From these measurements the difference of the work functions of tip and sample ΔØ was obtained as a function of V_G (see Fig. 3). Starting at $V_G = 0$, $V_{\Delta Ø}$ is increased if V_G is lowered, *i.e.,* if the carrier density at the $LaAlO_3$–$SrTiO_3$ interface n is lowered. At $V_G = –70$ V, the slope of the $V_{\Delta Ø}(V_G)$ curve changes sign. $V_{\Delta Ø}$ displays a clear minimum at $V_G = –95$ V. Using a second sample and different tips, we found that this minimum was reproducible. The V_G value of the minimum was found to differ among the samples, which we attribute to differences in the response of the samples on electric fields. The shape and the composition of the tips used in the experiments affected the absolute value of the minimum. A decrease of ΔØ, i.e. a decrease of the work function of the sample $Ø_s$, corresponds to an increase of the sample‘s chemical potential μ_s. Our data show that $V_{\Delta Ø}$ decreases in the voltage range from –70 V to –95 V. Hence in this range $d\mu_s/dn < 0$, the electronic compressibility is negative.

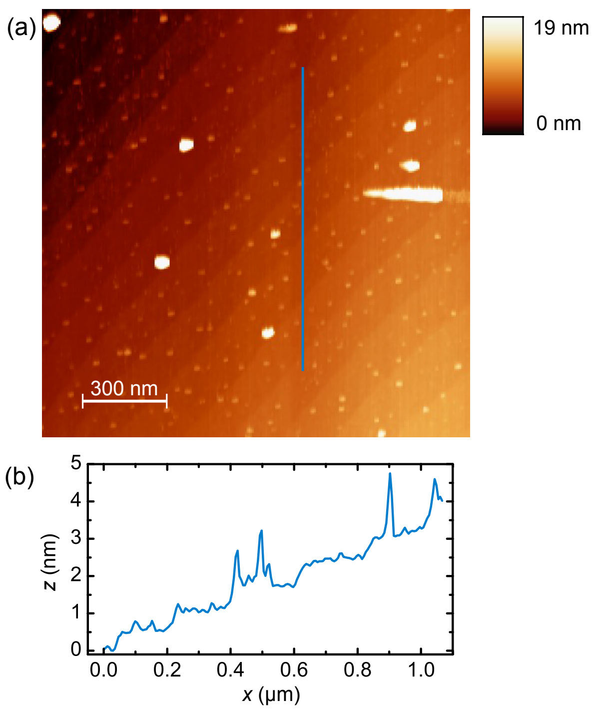

Figure 2: Topographic frequency-modulation scanning force microscopy image of the $LaAlO_3$ film acquired at 4.7 K. The image was recorded with a scanning speed of 75 nm/s and a frequency shift of −1.8 Hz. The free resonance frequency of the cantilever was 25 926.6 Hz, the quality factor was 20 320 and the oscillation amplitude was set to 3.4 Å. The white dots in the scan are caused by adsorbates on the sample surface. (b) Profile taken along the blue line plotted in (a) (from [3]).

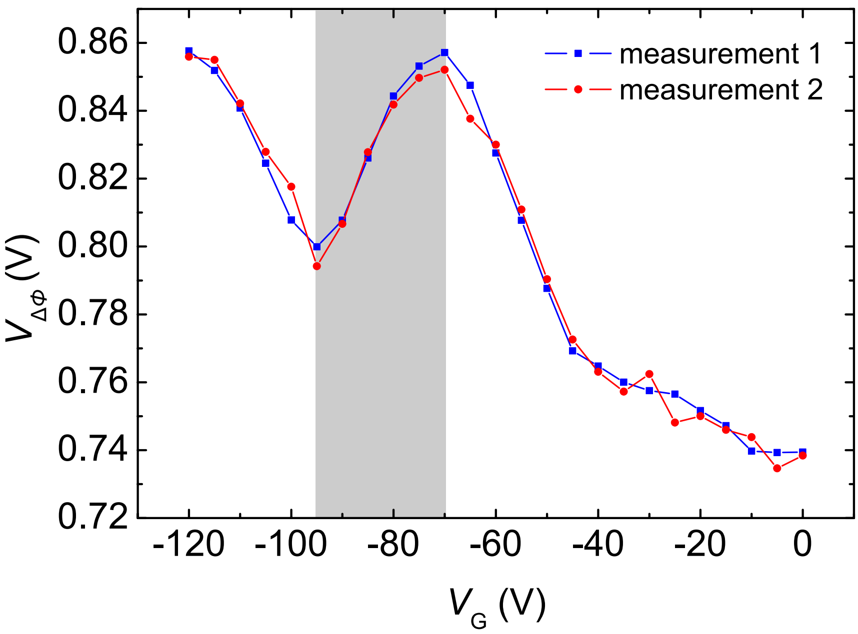

Figure 3: Contact potential difference $V_{\Delta\varnothing}$ measured as a function of V_G. The measurements were performed at 4.7 K. Spectrum 2 was recorded directly after spectrum 1. $V_{\Delta\varnothing}$ was obtained through quadratic fits of the Δf (V_{ts}) spectra (from [3]).

We have therefore shown with Kelvin probe microscopy that the conducting $LaAlO_3$–$SrTiO_3$ interface exhibits a negative electronic compressibility at low carrier densities even if the $LaAlO_3$ layer is only four unit cells thick [3]. This effect is consistent with the measurements reported in [2], which found a negative electronic compressibility close to the metal–insulator transition for ten and twelve-unit-cells-thick $LaAlO_3$ layers. The employed technique is independent of the previously used capacitance-measurement method. Moreover, as we deplete the interface solely by applying voltages to the back of the $SrTiO_3$ substrate, the use of a gate on top of the $LaAlO_3$ film is dispensable. It is therefore possible to measure samples with ultrathin $LaAlO_3$ films.

References:

[1] *Ohtomo, A. and H.Y. Hwang.* Nature **427**, 423–426 (2004).

[2] *Li, L., C. Richter, S. Paetel, T. Kopp, J. Mannhart, and R.C. Ashoori.* Science **332**, 825-828 (2011).

[3] *Tinkl, V., M. Breitschaft, C. Richter, and J. Mannhart.* Physical Review B **86**, 075116 (2012).

In collaboration with:

V. Tinkl, M. Breitschaft, and T. Kopp (Universität Augsburg)