

Supporting Information

Local inhomogeneities resolved by scanning probe techniques and their impact on 2DEG formation in oxide heterostructures

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S1: Line profiles of the topographic features of STO substrates

To exclude that the phase contrast is induced by the area in proximity to the terrace edges, topography line profiles of the STO substrates shown in the main paper are analysed here. The AFM image shown in Figure S1 a is an example of a fully TiO₂ terminated STO substrate (case [A] STO substrate). The scan was tilted to align the terrace planes with the image plane for better height comparison. Two line profiles are applied and shown in red and blue in Figure S1 b. From one plane to the next, a height difference of roughly 0.4 nm is observed which corresponds well to the expected lattice constant of STO (0.3905 nm). In comparison, the same scan as it was shown in Figure 2 b-d of the main paper is shown in Figure S1 c. The scan was tilted in the same manner as Figure S1 a and two lines profiles were inserted in red and blue which are shown in Figure S1 d. From one plane to the next, the same height difference is observed as present in Figure S1 b. However, the topographic features extending out of the terrace edge show an intermediate step between the two planes. The step height of the intermediate feature is close to half a unit cell as would be expected from a change in surface termination. Evaluation of the exact step height, however, is close to the topography resolution limit and is moreover impeded by potential surface adsorbates such as Sr(OH)₂.^[1] Therefore, these results support our interpretation of the topographic features being a SrO termination of the STO single crystal. However, for the evaluation of the complete areas and later comparison to conductivity and optical maps, the mapping of SrO terminated areas is more sufficient. For this reason, the AFM phase contrast was focused on in the main paper.

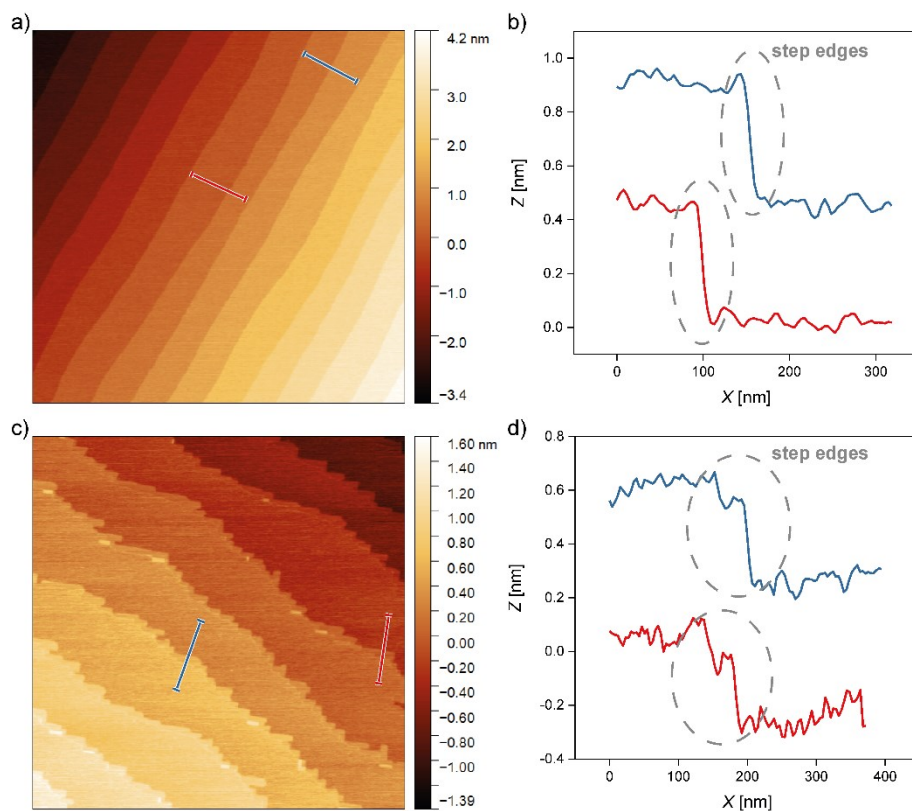


Figure S1: AFM images of STO substrates including line profiles are shown. a) Shows a case [A] (TiO_2 termination) STO substrate with two line profiles (red and blue), which are shown in b). c) Shows a case [B] (mixed termination) STO substrate with two line profiles (red and blue), which are shown in d). The step edges in the line profiles are circled by dashed lines for clarity. The AFM scans are $2 \times 2 \mu\text{m}$ in size.

S2: Cross-sectional line profiles of the phase contrast in AFM measurements

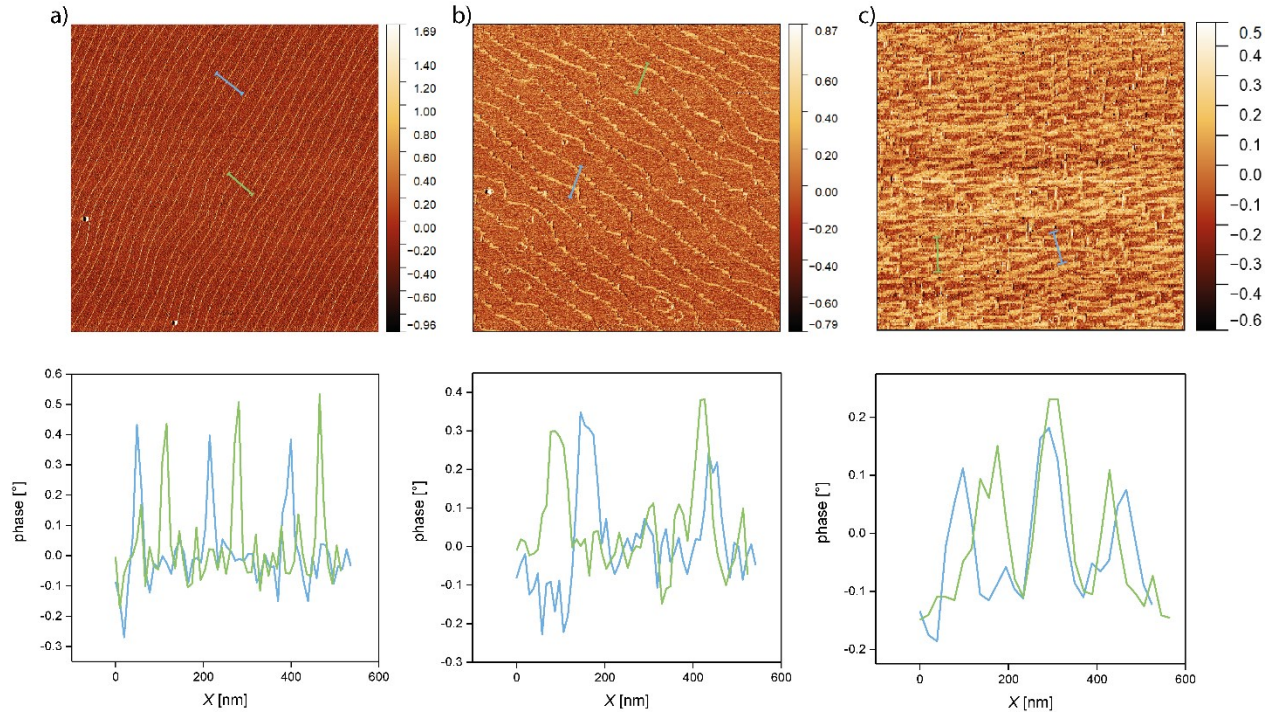


Figure S2: The phase contrast images of STO substrates shown in Figure 2 e-g of the main paper are shown with cross sectional line profiles applied in green and blue. The phase images are shown in the top row while the line profiles are shown in the bottom row. a) shows the result obtained from a case [A] STO substrate, b) for a case [B] STO substrate and c) for a case [C] STO substrate. The AFM scans are $5 \times 5 \mu\text{m}$ in size.

To prove that the phase contrast in AFM measurements on STO substrates is robustly observed, cross-sectional line profiles are shown in Figure S2. The shown phase images are identical to the main paper (Figure 2 e-g) with cross-sectional line profiles applied in the regions marked in green and blue. These line profiles are shown beneath the phase images. For a case [A] STO substrate (Figure S2 a), the phase contrast is present as sharp lines across the phase image. This behavior is also present in the line profile, where the phase sharply increases. The phase contrast is broadened for case [B] STO substrates (Figure S2 b) which is visible in both the phase image and in the line profile, where the heightened phase persists over a larger distance. An additional broadening of the phase contrast is observed for case [C] STO substrates (Figure S2 c), where the phase is heightened over an even wider area. Therefore, the presence of bigger areas with heightened phase contrast could be verified by the cross-sectional line profiles.

S3: Pulsed Laser Deposition

All LAO thin films shown in the original manuscript were grown in the same PLD chamber under the same conditions (see experimental details). In Figure S3 a summary of thin film growth of a 4 uc LAO/STO sample on a substrate of mixed termination is shown. The intensity of the primary reflected RHEED spot is plotted over time in Figure S3 a and shows clear oscillations from beginning to end of the deposition. The STO substrate showed a typical RHEED pattern that is

presented in Figure S3 b and is still visible after deposition with an overall lowered intensity (Figure S3 c). The RHEED pattern shows diffracted spots to the left and right of the specular spot, which correspond to the in-plane lattice of the STO crystal and later of the LAO thin film. Between diffracted and specular spot, faint additional spots are observable, which correspond to a 2x2 reconstruction pattern. This reconstruction can be introduced by high amounts of Sr at the surface^[2] which aligns with the interpretation of phase contrast in AFM. The topography of the substrate, which can be seen in Figure S3 d, is qualitatively replicated by the LAO thin film (Figure S3 e). All in all, the LAO thin films deposited on substrates of mixed termination do not show discrepancies to the ones deposited on perfectly terminated STO.

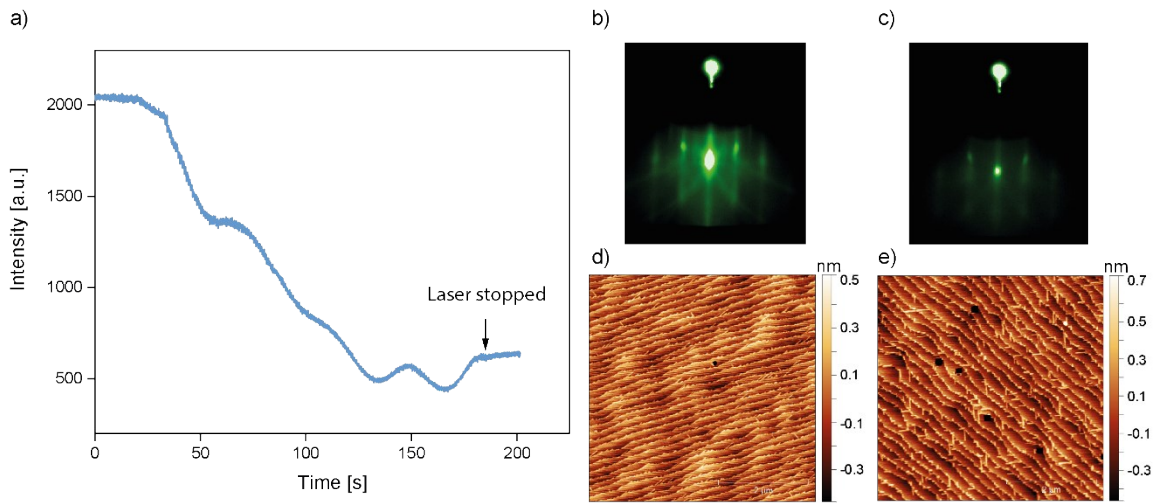


Figure S3: A summary of the LAO thin film growth is shown a) plots RHEED intensity of the primary reflected spot over time b) shows the RHEED pattern before deposition and c) after deposition d) shows the AFM measurement of the STO substrate and e) of the LAO thin film. The AFM scans are 5x5 μm in size.

S4: Proof of principle

In this experiment the ability of the c-AFM setup to detect currents through the 2DEG of the LAO/STO sample was tested. For this purpose, a LAO/STO sample was first measured in c-AFM without side contact. The result can be seen in Figure S4 a, which shows the recorded topography in the top image and the local current map in the bottom image. No currents above noise level could be measured in this configuration. In the second step, a side electrode using silver paste was applied to the sample and the measurement was repeated. Note that after such *ex-situ* steps, the sample was heated inside a preparation chamber at 200 °C for 30 min to desorb any surface contaminations that may be present due to air exposure. The result is shown in Figure S4 b, where now a significant enhancement of measurable current is visible. Removing the side electrode in the third step (Figure S4 c) leads to a vanishing of measurable currents. Through this series of

experiments, it can be deduced that current is indeed coupled into the 2DEG of the LAO/STO sample and therefore depends in its magnitude on its local electrical resistance.

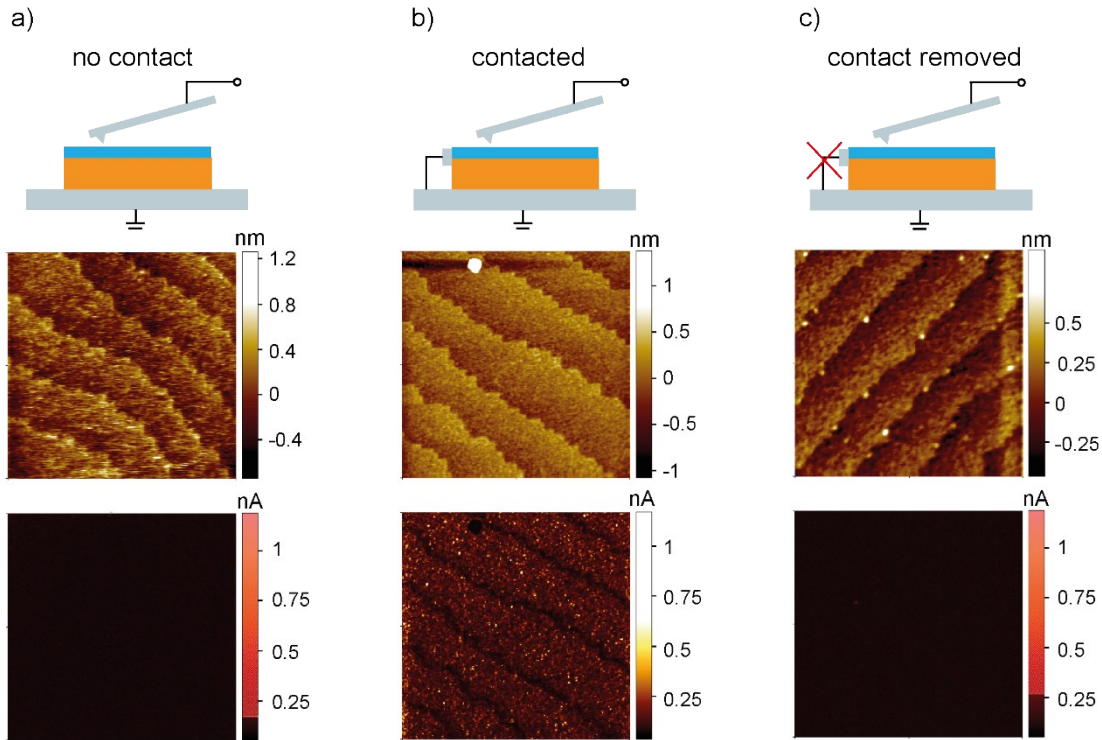


Figure S4: A consecutive series of c-AFM measurements on a LAO/STO sample are shown with a) the sample contacted at the backside of the substrate b) the sample contacted through a side electrode and c) after removal of the side electrode. The AFM scans are $1 \times 1 \mu\text{m}$ in size.

S5: Cross-sectional line profiles of the c-AFM current maps

To further analyze how the dark regions, measured in c-AFM widen from case [A] to case [C] LAO/STO samples, cross-sectional line profiles are shown in Figure S5. In all cases the same current maps are shown as in the main paper. The green and blue line profiles were drawn in a way to position the terrace edge roughly at the same position. For a case [A] LAO/STO sample (Figure S5 a) the terrace edge is almost indiscernible in the line profiles, showing only a sharp feature of lower current. The line profiles of the case [B] LAO/STO sample (Figure S5 b) show a much more pronounced and wider trench of the current right at the position of the terrace edge. The trench is further widened for the case [C] LAO/STO sample (Figure S5 c), extending about 50 nm at the scanned position. These line profiles prove that the contrast observed on LAO/STO samples in c-AFM robustly present, despite the overall low currents.

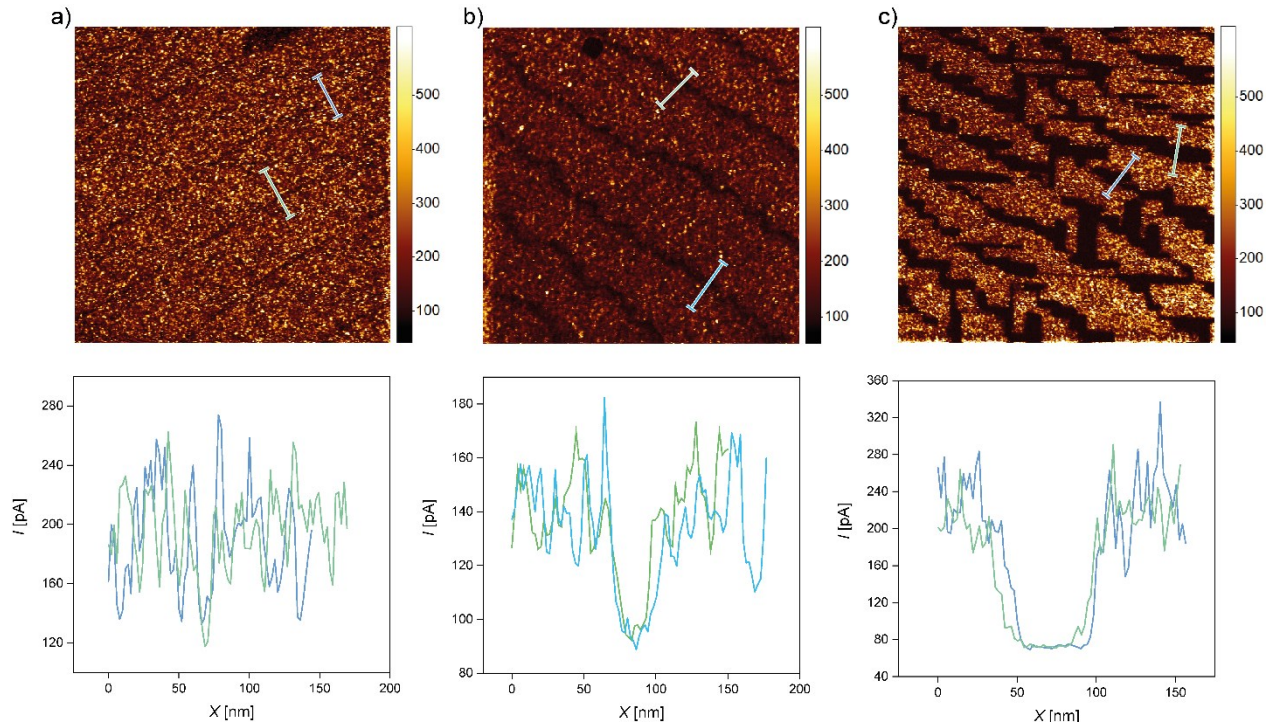


Figure S5: Current maps of c-AFM measurements on LAO/STO samples are shown (cf. Figure 3 of the main paper) with cross-sectional line profiles applied as marked by the blue and green inserts. Current maps are shown in the top row while the corresponding line profiles are shown in the bottom row. a) Shows the measurement for a case [A] LAO/STO sample, b) for a case [B] LAO/STO sample and c) for a case [C] LAO/STO sample. The AFM scans are $1 \times 1 \mu\text{m}$ in size.

S6: C-AFM measurements of thicker LAO/STO samples

In the main paper, c-AFM results are only shown for 4 uc LAO/STO, where high currents can be measured. To ensure that the contrasts recorded in c-AFM current maps is not caused by a varying thickness of the LAO thin film, measurements on thicker LAO/STO samples were conducted. The results for a 5 and 8 uc sample are shown in Figure S6 a and b respectively. For 5 uc LAO/STO (Figure S6 a), the same result as seen in the main paper is observed, where the topographic features induce a sharply resolved contrast in the current maps. In the case of 8 uc LAO/STO (Figure S6 b) the resolved contrast is much weaker and the applied voltage had to be increased to 6 V to induce measurable currents. Note that the absolute current range of 300 pA is close to the noise level. However, dark lines similar to the ones observed in Figure 3 b (main paper) are still observable. As the contrast in current maps is also present in samples with thicker LAO layers, it is not induced by a local thickness variation around 4 uc but has to be caused by another interface phenomenon.

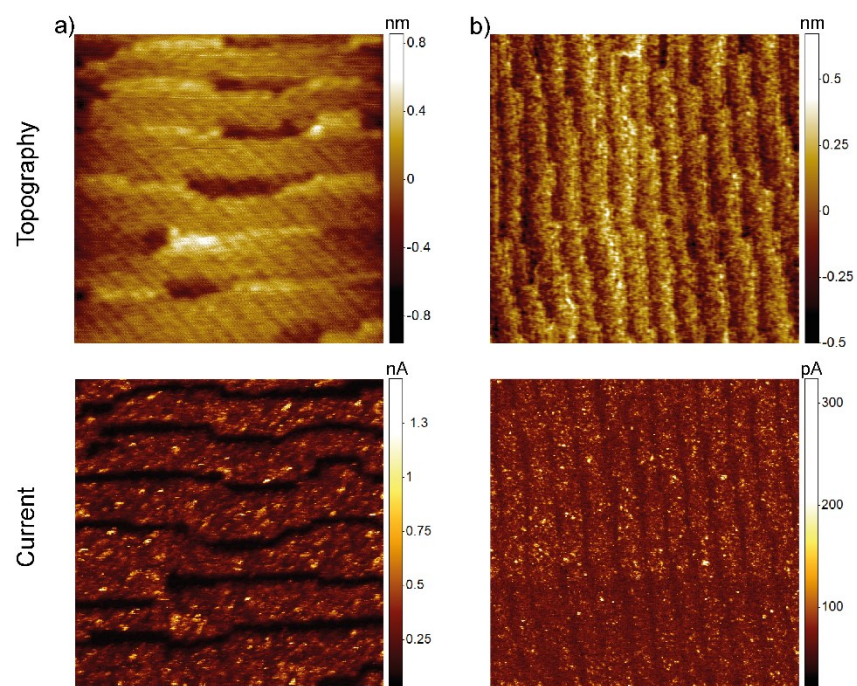


Figure S6: c-AFM measurements conducted on LAO/STO samples with mixed termination are shown with the topography in the top row and the current maps in the bottom row. a) Shows a 5 uc LAO/STO sample. b) Shows an 8 uc sample. The AFM scans are 1x1 μm in size.

S7: Current voltage characteristic of LAO/STO samples in c-AFM

Here the characteristic behavior of current to voltage ($I(V)$) for the measured currents of a 4 uc LAO on STO sample is shown in the used c-AFM system (Figure S7). For this, the tip was held at one position while the voltage amplitude was swept 10 times from -4 to +4 V and back. The curve is corrected to the offset of about 100 pA that is present in this setup (also in vacuum without a sample). In these measurements it can be seen that the sample does not show any significant change in its $I(V)$ behavior over the whole experiment, except for a slight hysteresis of the current. Furthermore, it can be shown that significant currents can be detected, which do not lead to an alteration of sample properties. The $I(V)$ curve shows a symmetric behavior, hinting at a tunneling mechanism. To investigate the nature of the tunneling mechanism, the characteristic plots for direct tunneling ($J \propto V$), Fowler-Nordheim ($\ln(J/V^2) \propto 1/V$) and Poole-Frenkel emission ($\ln(J) \propto V^{1/2}$) are shown in Figure S8. For this analysis, first the average of the $I(V)$ voltage sweeps was determined to minimize the noise level of the measurement. The result is shown in standard linear form in Figure S8a and in logarithmic form in Figure S8b. The noise level is sufficiently suppressed by the averaging. The characteristic plot for Fowler-Nordheim emission is shown in Figure S8c in which no clear linearity can be seen over the used voltage range, leading to the conclusion that it is not the dominant contribution to the $I(V)$ curve. The characteristic plot for Poole-Frenkel

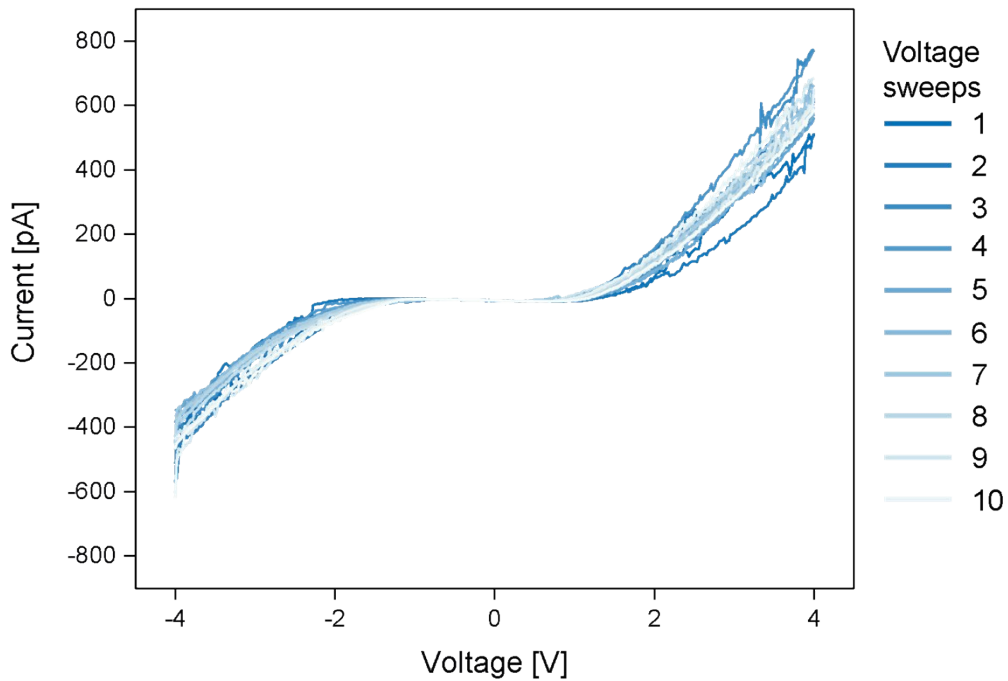


Figure S7: The measured current over applied voltage is shown for a sample of 4 uc LAO on STO in the c-AFM setup used. The colors from deep to light blue each represent a consecutive voltage sweep.

emission is shown in Figure S8d, where a linearity is visible towards high voltages. For high voltages the $I(V)$ characteristic can be therefore described by a Poole-Frenkel emission, which requires the existence of trap states inside the LAO thin film. This transport mechanism was already proposed by Swart et al.^[3] for the transport across Co/LaAlO₃/SrTiO₃ heterojunctions. The complete $I(V)$ curve, however, cannot be described by one single transport mechanism. Most probably a convolution of multiple conduction mechanisms is taking place, making the exact determination more complicated. An electrically induced ionic motion such as migration of oxygen vacancies inside the STO, could cause the observed slight hysteresis in the $I(V)$ curves. However, such a mechanism cannot explain the complete observed behavior. For one, the same contrast is observed in s-SNOM measurements, where no electrical field is applied (see Figure 4). An electrically induced behavior should not be reproducible by this method. Second, no contrast is observed below the critical thickness for 2DEG formation. Therefore, the observed current cannot be a consequence of ionic motion alone. For a better understanding of the exact transport phenomena, temperature dependent measurements would be needed. In summary, a convoluted transport is observed that can be described by a trap assisted tunneling process at high voltages.

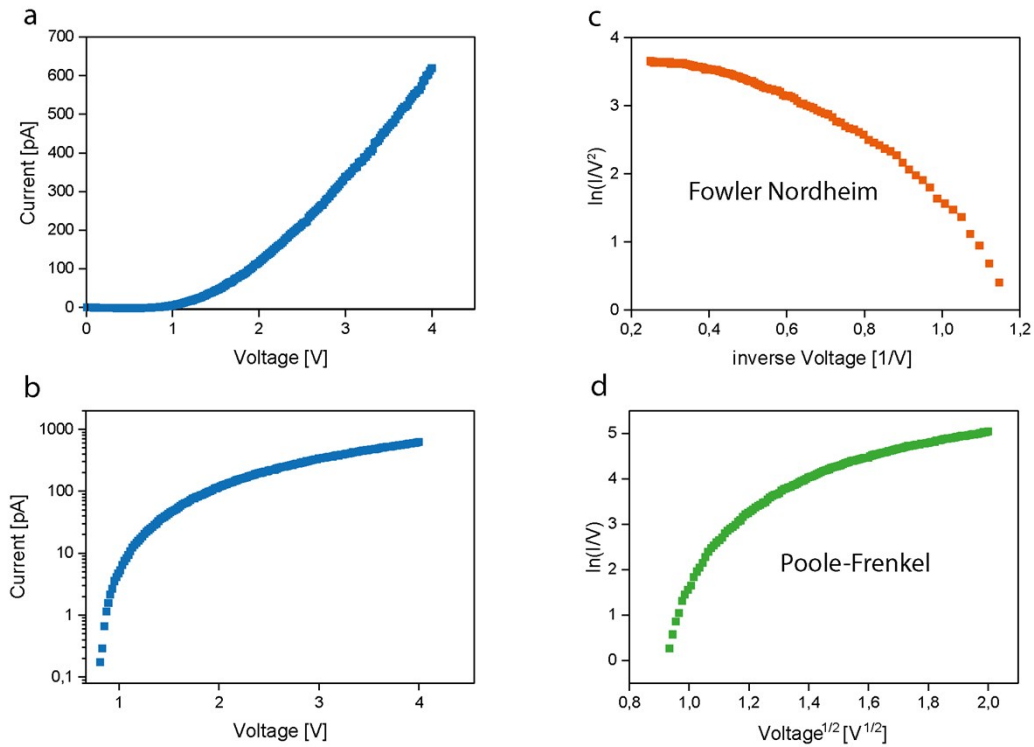


Figure S8: a) shows the current average resulting of ten voltage sweeps, b) shows a) on a logarithmic scale, c) shows the $\ln(I/V^2)$ over $1/V$ that is characteristic for Fowler-Nordheim emission d) shows $\ln(I/V)$ over the square root of V which is characteristic for Poole-Frenkel emission

S8: s-SNOM contrast dependence on tip-sample distance; histogram of s_2

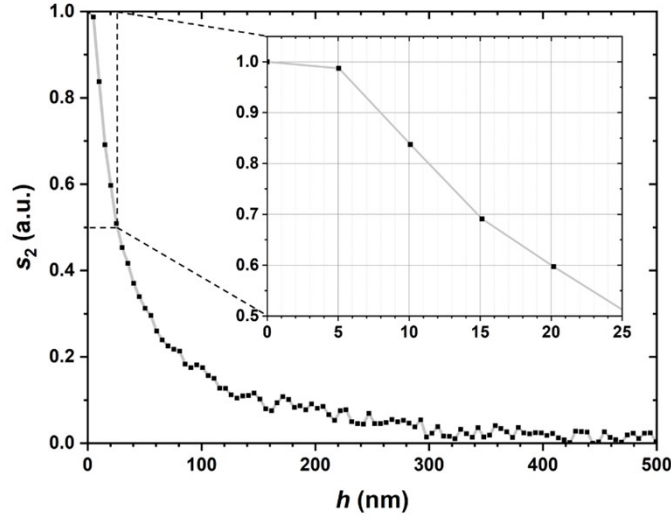


Figure S9: The retraction curve of the optical near-field measured on a LAO/STO sample (normalized to $[0,1]$), i.e. signal as a function of tip-sample distance h is shown. The inset shows a zoom-in to the first 25 nm, with vertical lines in steps of 1 nm.

A retraction curve of the optical near-field signal s_2 measured on a LAO/STO sample is shown in Figure S9 as a function of tip-sample distance h and normalized to $[0,1]$. The optical near-field signal decays exponentially with a $1/e$ decay length of 40 nm in this measurement, which roughly corresponds to the tip radius. The inset presents a zoom-in of the first 25 nm, clearly indicating that the signal varies

only 0.1% for a 0.4 nm change in tip-sample distance. Taking into account the sparsity of data, even in the region

of steepest slope the signal variation over $\Delta h = 0.4$ nm is only 1.4%. Thus, we conclude that the observed contrast at the step edges is not a topographic artifact.

To quantify the measured contrast observed at the measured LAO/STO sample, a line profile is presented in the main text (Figure 4d), which clearly shows signal variations across the step edges. Due to the relatively blunt optical tip of approximately 40 nm radius, the near-field signal is averaged across conducting and non-conducting regions (30-60 nm, as determined by c-AFM line profiles in section S5). As a result, intermediate values are recorded instead of a binary distribution of two distinct states, leading to reduced contrast. The histogram (Figure S10a) of the s_2 -signal for the measurement (cf. Figure 4c of the main text) thus shows only a single peak. Using a mask based on manual extraction of topographic steps (Figure S10b), a separation of different s_2 signal levels is possible (Figure S10c). Using this method, a signal contrast of 4% between step edges and faces can be determined as a lower bound. Thus, the evaluation of line profiles as presented in the main text gives a better estimate for the expected contrast between barely-resolved features compared to histogram analysis.

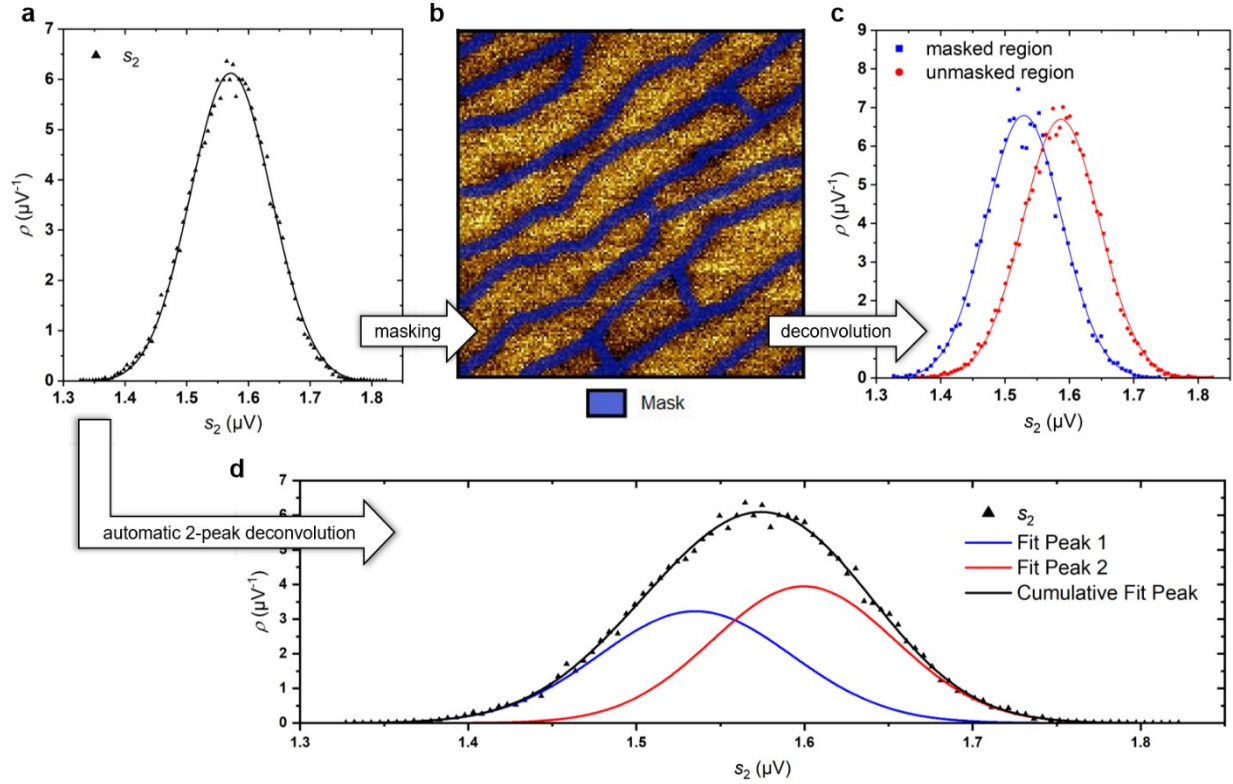


Figure S10: Evaluation of s-SNOM contrast on LAO/STO using histograms. a) Histogram of the s_2 -signal showing that intermediate values are prevalent. b) Mask based on manual extraction of topographic steps, overlaid onto the s_2 channel (cf. Figure 4c in the main text, image size $1 \mu\text{m} \times 1 \mu\text{m}$). c) Histograms of s_2 -signal in regions of the step edges (masked, blue) and on the step faces (unmasked, red), with respective Gaussian fits. The center positions are $s_{2,\text{edge}} = 1.53 \mu\text{V}$ and $s_{2,\text{face}} = 1.59 \mu\text{V}$, resulting in a lower bound of 4% for the signal contrast. d) Separation of step edges (blue) and step faces (red) using a 2-peak deconvolution routine. The center positions are $s_{2,\text{edge}} = 1.54 \mu\text{V}$ and $s_{2,\text{face}} = 1.60 \mu\text{V}$, also resulting in a lower bound of 4%.

S9: SNOM contrast dependence on electronic properties of LAO/STO

In Figure 4 of the main text, a contrast between step edges and faces is shown in the near-field optical signal, which is discussed as a result of diminished interfacial 2DEG. In s-SNOM, the electronic properties of the sample and the detected signal are linked via the sample dielectric function, and this relation is often not trivial. Therefore, theoretical investigations are necessary to evaluate which influence on the measured signal can be expected for changes in charge carrier density n . Additionally, the dependence on electron mobility is modelled, to determine whether a reduced 2DEG mobility could explain the observed contrast as well. Finally, the two influences are compared directly.

To model the s-SNOM contrast, the Finite Dipole Model (FDM)^[4] was employed in combination with a Transfer Matrix Method (TMM), resulting in a powerful predictive tool^[5] to simulate the near-field response of arbitrarily layered systems with known dielectric function. SNOM contrast simulations of the LAO/STO 2DEG were already published^[6] and are utilized here with averaged

values obtained from Hall measurements: $n_{2D} = 9.96 \times 10^{13} \text{ cm}^{-2}$, $\mu = 4.09 \text{ cm}^2/\text{Vs}$. In order to obtain a volume concentration, the sheet carrier concentration is distributed over a thickness of $d_{2DEG} = 7.5 \text{ nm}$, resulting in a volume concentration of $n_{3D} = 1.33 \times 10^{20} \text{ cm}^{-3}$ (vertical red line in Figure S11c). To present a realistic frame of reference, we take into account non-averaging behaviour due to the insulating step edges and the fact that the 2DEG might be confined to a smaller thickness, yielding higher charge carrier concentrations. As a result, we look at a variation between $n_{3D} = 1 \times 10^{20} \text{ cm}^{-3}$ and $6 \times 10^{20} \text{ cm}^{-3}$, with additional values below $1 \times 10^{20} \text{ cm}^{-3}$ added to bridge the gap to the insulating sample (cf. Figure S11c). Similar bounds can be established for the mobility, which is expected to stay below $\mu = 10 \text{ cm}^2/\text{Vs}$ at room temperature (cf. Figure S11f). Finally, it is important to note that the 2DEG is located within the STO side and is therefore modeled as free electrons (Drude contribution) on a background given by STO phonons (cf. Eq S1). As such, the dielectric function of the 2DEG layer is identical to that of the STO substrate, as long as no free charge carriers are present.

$$\varepsilon(\omega) = \varepsilon_{STO} + \varepsilon_{Drude} = \varepsilon_{STO} - \frac{\omega_p^2}{\omega^2 + \gamma^2} + i \frac{\gamma \omega_p^2}{\omega(\omega^2 + \gamma^2)} \quad (\text{Eq. S1})$$

In the Drude formalism, the plasma frequency ω_p depends on n , while the damping factor γ depends inversely on μ . Figure S11a-c show the dielectric function of the system and resulting SNOM contrast when adding a 2DEG of (volume) charge carrier concentration n to a formerly insulating LAO/STO sample, starting with changes to the dielectric function of pure STO (black) in Figures S11a (real part) and S11b (imaginary part). Here, a rising n is marked by a transition to red color (cf. Figure S11c for explicit values of n). Using these dielectric functions for the 2DEG layer, the s-SNOM contrast of LAO/STO samples is presented in Figure S11c, showing the s-SNOM amplitude as a function of n . A strong increase in signal can be seen for high values of n , while two minima can be observed around the nominal charge carrier density of $1.33 \times 10^{20} \text{ cm}^{-3}$ (vertical red line in Figure S11c).

In Figure S11d-f, the influence of electron mobility μ on dielectric function (Figure S11d, e) and s-SNOM contrast (Figure S11f) is shown. A major difference to the behavior with n is that the zero crossing of $\text{Re}[\varepsilon]$ converges for higher values of μ (around 1150 cm^{-1} in Figure S11d) and that the imaginary part at the measurement frequency (vertical grey line in Figure S11e) first rises until $\mu = 3.5 \text{ cm}^2/\text{Vs}$ and is then reduced for higher mobilities. As a result, the influence of μ on the s-SNOM contrast is different to the influence of n , e.g. high μ -values decrease the signal slightly while high n -values increase it strongly.

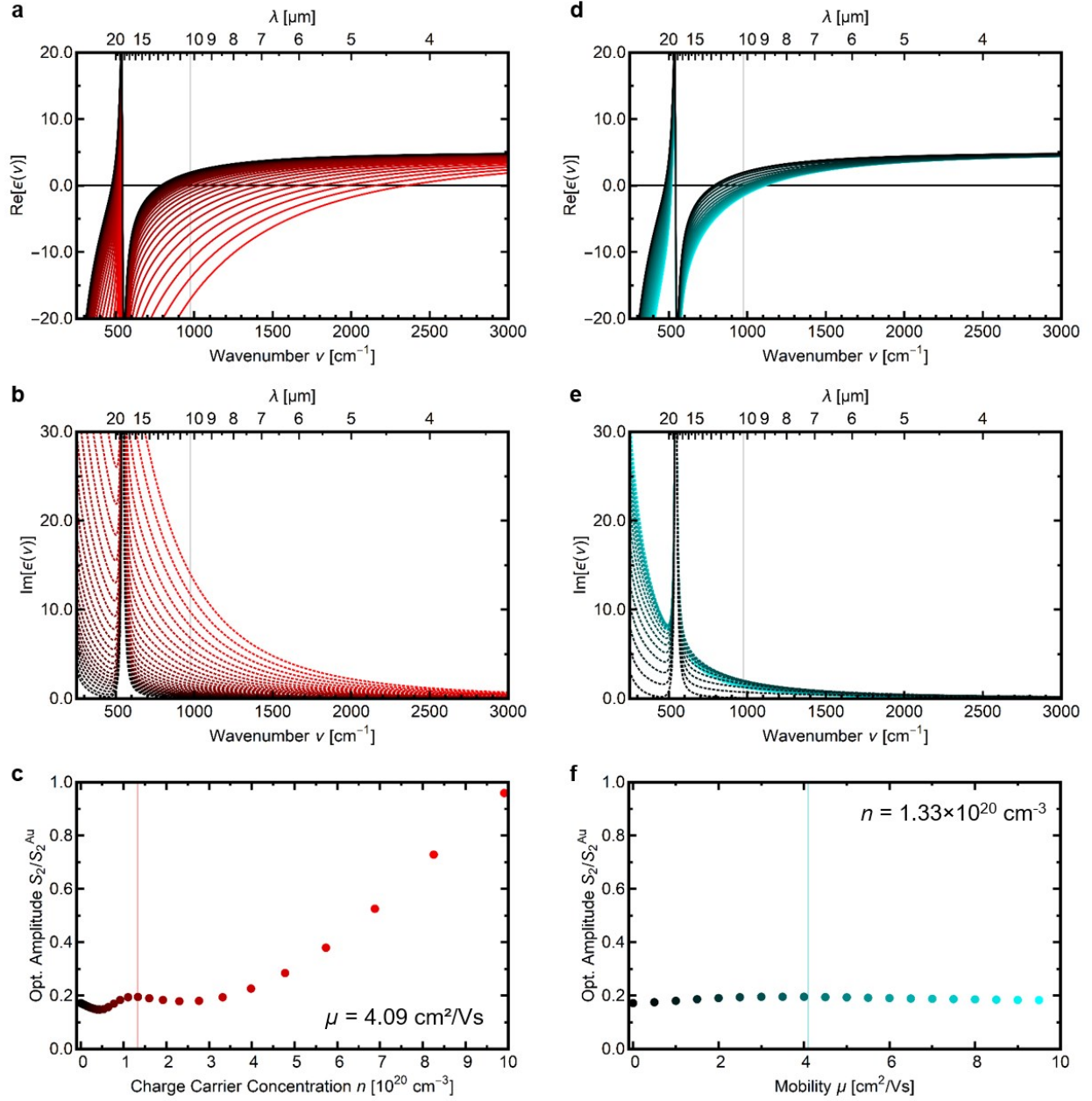


Figure S11: Simulation of s-SNOM contrast dependence on charge carrier concentration and mobility in LAO/STO: **a** real part of the dielectric function $\text{Re}[\epsilon]$, showing the phonon resonance of pure STO (black) and the addition of a 2DEG with rising charge carrier concentration n (transition to red, values shown in c). The zero-crossing shifts to higher frequencies with rising n ; the vertical grey line indicates the measurement frequency of $\nu = 974 \text{ cm}^{-1}$. **b** corresponding imaginary part $\text{Im}[\epsilon]$, showing an overall increase with rising n . **c** Simulation of the normalized s-SNOM amplitude at the measurement frequency of $\nu = 974 \text{ cm}^{-1}$ as a function of n . The vertical red line indicates the value determined by Hall measurements. **d** $\text{Re}[\epsilon]$ showing the influence of a 2DEG with rising mobility μ (transition to cyan, values shown in f). **e** corresponding imaginary part $\text{Im}[\epsilon]$, showing first an increase and then a decrease with rising μ . **f** Simulation of the normalized s-SNOM amplitude at the measurement frequency of $\nu = 974 \text{ cm}^{-1}$ as a function of μ . The vertical cyan line indicates the value determined by Hall measurements.

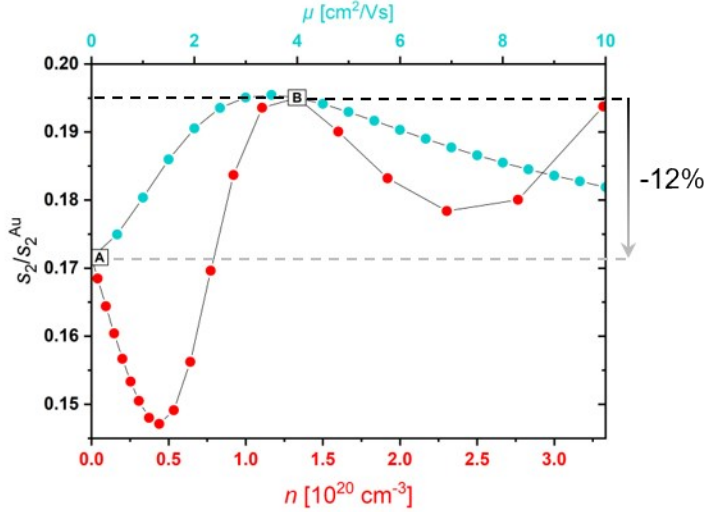


Figure S12: zoom-in of s-SNOM contrast dependence on charge carrier density (red) and mobility (cyan) in LAO/STO, with values identical to Figures S6c and f. Two points are common to both curves: **A** is the contrast of the STO substrate, which is identical to $n = 0$ in the red curve and to $\mu = 0$ in the cyan curve; **B** is the investigated sample, with $n = 1.33 \times 10^{20} \text{ cm}^{-3}$ and $\mu = 4.09 \text{ cm}^2/\text{Vs}$ in both curves. Dashed horizontal lines indicate the difference in contrast between regions with and without electrons, respectively.

Figure S12 presents a zoom-in of the simulated s-SNOM contrast as shown in Figures S11c and f. Here, the distinct influence of both parameters is clearly visible: while $s_2(\mu)$ has a maximum at $3.5 \text{ cm}^2/\text{Vs}$ and drops off to both lower and higher mobilities, $s_2(n)$ additionally has two minima around 0.5 and $2.5 \times 10^{20} \text{ cm}^{-3}$. Point **A** marks LAO/STO without free charge carriers and **B** the investigated sample with 2DEG. The contrast between the two cases is predicted to be 12% and could be achieved by

both a reduced charge carrier concentration or mobility. While the quantitative results depend slightly on several factors, such as the assumed 2DEG thickness, the overall qualitative result is nonetheless persistent with a reduced signal in regions without charge carriers (**A**) and a contrast of ~10% between **A** and **B**.

S10: Low-Temperature Hall analysis

Here the Hall measurements conducted in low temperatures are shown in Figure S13 for a case [A] and [C] LAO/STO sample. The data was recorded using a mimicked Hall bar geometry where contacts were achieved by Al wire bonding. A sketch of the bonding scheme is shown as inset in Figure S13 a. A rectangular piece of roughly $1.5 \times 5 \text{ mm}^2$ was cut from the $5 \times 5 \text{ mm}^2$ sample pieces. Then rows of densely bonded wires were applied at both ends of the piece to ensure an evenly distributed current through the sample. The wires measuring magnetoresistance (V_{MR}) and the Hall voltage (V_{H}) are applied in current direction and perpendicular to it. All shown data was furthermore symmetrized to compensate offset voltages.^[7] In the temperature regime below 100 K the linear Hall effect observed in LAO/STO at room temperature becomes non-linear. This behavior results from the two available conduction bands that cannot be distinguished at higher temperatures due to phonon scattering. The carrier populations (n_1, n_2) and mobilities (μ_1, μ_2) can be extracted from the Hall coefficient (R_{H}) under an applied magnetic field (B) by the use of a two carrier model that follows the expression of ^[8]

$$R_H = - \frac{1}{e} \frac{\left(\frac{n_1 \mu_1^2}{1 + \mu_1^2 B^2} + \frac{n_2 \mu_2^2}{1 + \mu_2^2 B^2} \right)}{\left(\frac{n_1 \mu_1^2}{1 + \mu_1^2 B^2} + \frac{n_2 \mu_2^2}{1 + \mu_2^2 B^2} \right)^2 + \left(\frac{n_1 \mu_1^2}{1 + \mu_1^2 B^2} + \frac{n_2 \mu_2^2}{1 + \mu_2^2 B^2} \right)^2 B^2}$$

In Figure S13 a) the Hall coefficients (R_H) of a case [A] (blue) and [C] (orange) sample are shown over the applied magnetic field at a temperature of 5 K. The fit of R_H (red) is only applied in the range between 3 and 9 T as in the middle region an anomalous Hall effect additionally alters the shape of the two carrier model. It is more strongly pronounced for the case [C] LAO/STO sample than for case [A], but the higher overall resistance leads to additional noise on the measurement, making a clear conclusion more difficult. The anomalous Hall effect is generally an indication of magnetic moments which suggests that the LAO/STO heterostructures of mixed termination show a stronger magnetic signature. However, as the situation at these interfaces is rather complicated with multiple percolating 2DEGs, further investigations are needed to make this claim. In Figure S13 b) the extracted carrier densities are shown for the applied model over temperature. Both carrier populations are lower for the case [C] LAO/STO sample in respect to case [A]: The one of higher density (n_1), corresponding to a low mobility (μ_1 in Figure S13 c), by only a small amount while the carrier population of lower density (n_2), corresponding to a high mobility (μ_2 in Figure S13 c), by almost an order of magnitude. Qualitatively this can already be seen by the overall higher magnitude of R_H in case [C]. Also both mobilities are lower in the case [C] LAO/STO sample, however both carrier types seem to be lower by roughly the same factor in this case. In summary both carrier densities as well as their mobilities are lowered when SrO is present at the LAO/STO interface.

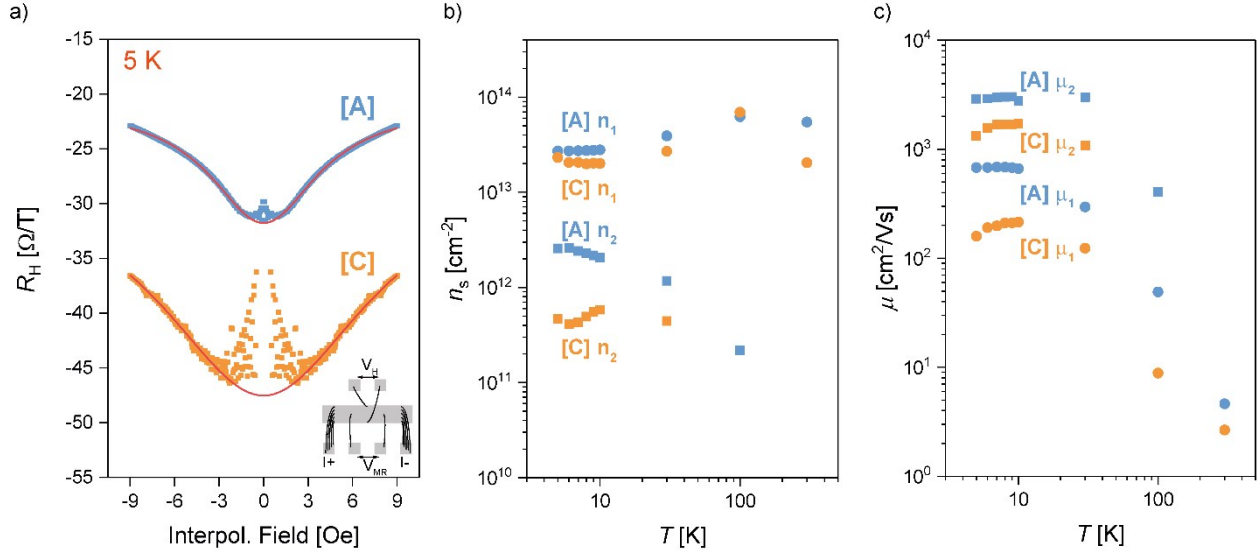


Figure S13: a) The Hall resistances R_{xy} over applied magnetic field for a case [A] (blue) and [C] LAO/STO sample (orange) are shown at a temperature of 5 K. A sketch of the used bonding scheme to achieve the mimicked Hall bar structure is shown as inset. b) The extracted carrier densities n_1 (dots) and n_2 (squares) over temperature are shown for a case [A] (blue) and [C] sample (orange). c) The extracted mobilities μ_1 (dots) and μ_2 (squares) over temperature are shown for a case [A] (blue) and [C] sample (orange).

Literature:

- [1] F. V. Hensling, C. Baeumer, M. Rose, F. Gunkel and R. Dittmann, SrTiO₃ termination control: a method to tailor the oxygen exchange kinetics, *Mater. Res. Lett.* **8**, 31-40 (2020)
- [2] S. Gerhold, Z. Wang, M. Schmid and U. Diebold, Stoichiometry-driven switching between surface reconstructions on SrTiO₃(001), *Surf. Sci.* **621**, L1-L4 (2014)
- [3] A. G. Swartz, S. Harashima, Y. Xie, D. Lu, B. Kim, C. Bell, Y. Hikita and H. Y. Hwang, Spin-dependent transport across Co/LaAlO₃/SrTiO₃ heterojunctions, *Appl. Phys. Lett.* **105**, 032406 (2014)
- [4] A. Cvitkovic, N. Ocelic and R. Hillenbrand, Analytical model for quantitative prediction of material contrasts in scattering-type near-field optical microscopy, *Opt. Express* **15**, 8550-8565 (2007)
- [5] B. Hauer, Nano-optical mapping of permittivity contrasts and electronic properties at the surface and beneath, PhD thesis (2015)
- [6] J. Barnett, M.-A. Rose, G. Ulrich, M. Lewin, B. Kästner, A. Hoehl, R. Dittmann, F. Gunkel and T. Taubner, Phonon-Enhanced Near-Field Spectroscopy to Extract the Local Electronic

Properties of Buried 2D Electron Systems in Oxide Heterostructures, *Adv. Funct. Mater.* **30**, 2004767 (2020)

- [7] F. Gunkel, C. Bell, H. Inoue, B. Kim, A. G. Swartz, T. A. Merz, Y. Hikita, S. Harashima, H. K. Sato, M. Minohara, S. Hoffmann-Eifert, R. Dittmann and H. Y. Hwang, Defect Control of Conventional and Anomalous Electron Transport at Complex Oxide Interfaces, *Phys. Rev. X* **6**, 031035 (2016)
- [8] V. K. Guduru, A. McCollam, J. C. Maan, U. Zettler, S. Wenderich, M. K. Kruize, A. Brinkman, M. Huijben, G. Koster, D. H. A. Blank, G. Rijnders and H. Hilgenkamp, Multi-band Conduction Behavior at the Interfaces of $\text{LaAlO}_3/\text{SrTiO}_3$ Heterostructures, *J. Kor. Phys. Soc.*, **63**, 437-440 (2013)