Electronic Transport Properties of LaAlO$_3$/SrTiO$_3$ Heterostructures Under Hydrostatic Pressure

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Motivation

The effect of stress on materials has been known for a very long time. It was learned that applying stress on crystalline materials directly changes the distance between atoms which could lead to changes in crystal symmetry. Such modifications consequently alter the physical properties of the material. The effect of stress on physical properties of solids was most widely studied and applied in semiconductor physics, where in 1950 the deformation potential theory was developed by Shockley and Bardeen, relating strain to the conduction band energy shift [1]. Among the simplest applications of stress are epitaxial strain and hydrostatic pressure. The former is generated by depositing thin films of materials on a substrate with slightly different lattice parameters while hydrostatic pressure is achieved by subjecting a sample to equal pressure from all directions.

Epitaxial strain has been used to tune and to induce new properties in semiconducting thin films. For example, it has been used to enhance the electron mobility in silicon based devices, leading to an increase in their switching speed [2]. In strongly correlated transition-metal-oxides, the influence of stress on the electronic properties is expected to be much more pronounced due to the strong coupling of the conduction electrons to the crystal lattice. Even slight distortions of the unit cell may substantially influence crystal fields, orbital occupancy, polarization, and exchange coupling in these oxides [3]. For example, epitaxial strain was used to tune the magnetic properties in LaCoO$_3$ films [4]. Furthermore, SrTiO$_3$(STO) films have been driven ferroelectric with a $T_c$ of 293K [5].

Uniaxial and biaxial strain dominantly affects the symmetry of the unit cell, while keeping the unit-cell volume almost constant, which hinders one from investigating the changes of the electronic properties that are induced by volume changes of the unit cell. However, these changes are accessible and have been explored by applying hydrostatic pressure. For example, it has been used to induce superconductivity in hydrogen sulfide compounds, with transition temperature about 200 K [6]. Also, in high temperature superconducting cuprates, $T_c$ of 164K
was achieved by hydrostatic pressure of 15 GPa [7]. And more recently, hydrostatic pressure of 23 GPa was used to reduce the Curie temperature of SrRuO$_3$ films from 150 to 77 K [8].

The two dimensional (2D) correlated electron system forming at the interface between LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) exhibits symmetry induced splitting of the Ti-3d orbitals. That orbital splitting may be likely influenced by the interatomic distance of the atoms in STO substrate, which greatly influences the electronic properties of the system. In addition to that, the relative permittivity of STO is very sensitive to hydrostatic pressure, especially in the low temperature regime [9]. Hence, the electronic properties of LAO/STO heterostructures are expected to be very sensitive to hydrostatic pressure, motivating measurements of electronic transport under non-ambient conditions, i.e. high pressure and low temperature.
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Chapter 1

Introduction

Artificial heterostructures (AHS’s) and superlattices (SL’s) have been at the core of condensed matter physics research and device technology over the past decades. Semiconductor HS’s and SL’s formed the basis for many advances in science and electronic devices. An example is the formation of a 2-dimensional electron gas in III-V-compound semiconductor heterostructures and in metal-oxide-semiconductor field-effect transistors (MOSFET’s), which not only led to a breakthrough in device technology, but also represented the basis for the discovery of the quantum Hall effect by von Klitzing et al. [16]. Another example is the giant magneto-resistance in metallic SL’s composed of paramagnetic and ferromagnetic layers [17], which are now used as sensing elements in the reading heads of hard disk drives.

At the beginning of this century, numerous efforts have been made to engineer such HS’s and SL’s using transition metal oxide (TMO) compounds. TMO’s can be found in almost every possible electronic ground state such as metallic, insulating, superconducting, ferromagnetic, ferroelectric and more. Many of these compounds show complex phase diagrams with competing ground states due to the delicate interplay of spin-, charge-, orbital and lattice interactions of electrons. External perturbations such as strain, hydrostatic pressure, electric and magnetic fields can very effectively change the orbital ordering patterns of electrons in the 3d bands.

The technological advancements in thin film deposition methods for complex oxides such as Pulsed Laser Deposition (PLD) and Oxide Molecular Beam Epitaxy (MBE), together with the good chemical treatment of substrate and nearly perfect lattice match between many TMO’s, made the fabrication of such HS’s or SL’s a reality. It is expected that combining such TMO’s in HS’s or SL’s would give rise to novel quantum phenomena at their interfaces with properties and functionalities that are not observed in metals or semiconductors.
A prominent example of such heterostructures is the discovery of a highly mobile two-dimensional electron system (2DES) at the interface between the two band insulators LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) by Ohtomo and Hwang in 2004 [18]. To explain this discovery, they proposed the polar catastrophe hypothesis, which attributes the formation of the 2DES to the polar discontinuity found at the interface. LAO film layers, i.e. Al$^{3+}$O$_4^{2-}$ and La$^{3+}$O$_2^{2-}$, are charged -1 and +1 respectively, while STO alternates neutral Ti$^{4+}$O$_4^{2-}$ and Sr$^{2+}$O$^2^-$ layers. This polar discontinuity leads to an electric field within the LAO film and an electrostatic potential builds up with increasing LAO film thickness, diverging eventually to infinity. To avoid this catastrophe, the system undergoes electronic reconstruction, resulting in a half elementary charge per formula unit to be transferred from LAO surface to the interface, specifically, to the Ti 3d bands in the top layer of STO substrate. First principles density-functional-theory(DFT) [11] calculations show that in Ti-3d $t_{2g}$ bands, the $d_{xy}$ orbitals, which are degenerate with $d_{xz}$ and $d_{yz}$ in bulk STO, exhibit a drop in their energy of about 250 meV due to the constrained movement of electrons in the direction perpendicular to the sample surface. This drop in the energy of $d_{xy}$ orbitals causes them to become the first orbitals for electrons to occupy, leading to one-type-carrier-transport as long as they are the only occupied orbitals. X-ray spectroscopy has also confirmed the splitting of the usually degenerate Ti-3d $t_{2g}$ bands [19]. This electronic reconstruction not only helps mitigate the effect of the polar catastrophe, but also leads to a metallic interface with a theoretical sheet carrier density of 3.3x10$^{14}$ cm$^{-2}$, which is the value suggested by the polar catastrophe hypothesis, where half electron per unit cell resides at the interface. However, the sheet carrier density ($n_s$) reported by other groups lies well below this value [20, 21, 22]. As sheet carrier density increases, a Lifshitz transition takes place between $d_{xy}$ orbitals and the two-fold degenerate $d_{xz}$ and $d_{yz}$ orbitals at $n_c \approx 1.7x10^{13}$ cm$^{-2}$ [15] leading to two-type-carrier-transport for $n_s>n_c$.

Another possible explanation for the metallic conductivity at the LAO/STO interface is the free electrons left by oxygen vacancies created in the STO substrate during sample growth [23]. The root of this hypothesis comes from the observation that conductive LAO/STO samples are usually grown at low oxygen partial pressure($p$(O$_2$)<10$^{-3}$mbar), while samples grown at high oxygen partial pressure ($p$(O$_2$)>10$^{-2}$mbar)are usually insulating. Ionic intermixing has also been discussed as a source of electrons as well [24]. It is possible for the La$^{3+}$ ions to substitute the Sr$^{2+}$ ions in STO substrate leading to an electron-doping effect. However, it is doubtful that the extent of the intermixing in the system is high enough to generate such high sheet carrier density.
In addition to the metallic conductivity, the interface exhibits a two dimensional superconducting transition, with critical temperature ($T_C$) of about 250 mK [25]. This superconducting transition is found to be sensitive to the charge carrier density at the interface, which can be tuned by applying a back gate voltage to the sample [14]. This tuning allows increase and decrease of $n_s$ by applying positive and negative gate voltage and hence the mapping of the so called “superconducting dome”, i.e. $T_C$ versus $n_s$. The maximum $T_C$ achieved with gate-voltage amounts to about 310 mK.

In this thesis, I will present investigations of the electronic transport properties of LAO/STO heterostructures under hydrostatic pressure. The structure of this work is organised as follows:

In chapter 2 an overview over the basic properties of the constituent materials (LAO and STO) as well as a summary of some important experiments performed on LAO/STO heterostructures under ambient pressure are given. In addition, the different hypotheses that attempt to explain the metallic conductivity at the interface are discussed.

The first part of chapter 3 deals with sample preparation processes, including substrate treatment, thin film growth using PLD as well as the optimization of the growth parameters. After that, the structural properties of the film and the sheet resistance dependence on film thickness are discussed. The final part describes the experimental apparatus and techniques used in order to perform transport measurements under hydrostatic pressure. In chapter 4, the effect of oxygen partial pressure used during growth on the electronic-transport properties of LAO/STO heterostructures at ambient pressure is presented and discussed.

The main part of this work is presented in chapter 5 where the electronic transport properties of LAO/STO heterostructures are investigated under different hydrostatic pressure, ranging from ambient to 1.64 GPa. First, the sheet resistance dependence on hydrostatic pressure is presented and discussed. The second part includes the effect of hydrostatic pressure on the sheet carrier density and Hall mobility. The third part of chapter 5 reports on the magneto transport properties, where magnetoresistance measurements are performed at different temperatures and hydrostatic pressures. The fourth and final part of this chapter deals with the role of electron-electron interaction and weak-localization at temperatures below 10 K. Chapter 6 gives a summary of the work and the results.
Chapter 2

Physical Properties of 
LaAlO$_3$/SrTiO$_3$ heterostructures 
under ambient conditions

2.1 Structural and dielectric properties of the constituents LAO and STO

2.1.1 Structural properties

LAO and STO are non-magnetic band insulators, with band gaps of 5.6 eV and 3.2 eV respectively [26, 27]. Both have crystal structure that belongs to the perovskite family (see Fig.2.1), with a pseudo cubic lattice constant of 3.905 Å for STO [28], and 3.79 Å for LAO [29]. At room temperature and ambient conditions STO displays a cubic structure with Pm3m (221) space group symmetry, whereas LAO shows rhombohedric structure with R-3ch (167) symmetry.
Perovskite (ABO$_3$) thin films are composed of layers of AO and BO$_2$, which alternate along the 001 direction. In case of STO, the two layers are Ti$^{4+}$O$_2$$^-$ and Sr$^{2+}$O$_2$$^-$, with a net charge of zero. For LAO, the two layers are Al$^{3+}$O$_2$$^-$ and La$^{3+}$O$_2$$^-$, with net charges of (-e) and (+e) respectively. The close lattice matching of TMO’s makes LAO and STO as widely used substrates for the growth of oxide thin films. One of the advantages of using STO as a substrate is the possibility to create a single terminated surface, i.e., TiO$_2$ or SrO [30, 31]. The treatment process (explained in detail in the next chapter) is of great importance in creating metallic LAO/STO heterostructures. The type of the interface and its physical properties are defined by the type of the substrate termination of the STO substrate. In case of TiO$_2$ termination (TiO$_2$-STO), the growth of LAO films starts with a LaO layer, leading to an n-type metallic LaO/TiO$_2$ interface. As for SrO terminated surface, the growth of LAO starts with an AlO, leading to an insulating p-type AlO/SrO interface. Schematic of the layered structure of a LAO/STO heterostructure with n-type interface (LaO/TiO$_2$) is shown in Fig. 2.2.
Due to the difference between LAO and STO lattice constants, LAO films grown on STO experience a tensile strain. The in-plane lattice mismatch $\varepsilon$ can be calculated by:

$$\varepsilon = \frac{a_S - a_F}{a_F} \times 100\%$$  \hspace{1cm} (2.1)

$a_S$ is the lattice constant of the substrate and $a_F$ is the bulk lattice parameters of the film material. For LAO on STO, this results in $\varepsilon = 3.034\%$

### 2.1.2 Dielectric properties

The STO dielectric properties are important and therefore will be reviewed before investigating the metallic interface properties of LAO/STO heterostructures. The reason is that the carriers responsible for the conductivity in LAO/STO heterostructures reside in Ti-3d orbitals of the STO substrate top layer, which means the interface properties are affected by STO dielectric properties. In this section we will review the dielectric properties of STO, and their dependence on temperature, pressure and electric field. Wang et al. [9] have studied the effect of temperature...
and hydrostatic pressure on relative permittivity of STO. The result is shown in Fig.2.3.

![Figure 2.3: Temperature dependence of relative permittivity ($\varepsilon_r$) of STO at different hydrostatic pressure. The figure is taken from Ref. [9]. The transition temperature $T_q$ to a quantum paraelectric state is indicated by arrow. $T_q$ increases with increasing $p$, see inset.]

The relative permittivity $\varepsilon_r$ increases with decreasing temperature. The increase is maximum at lower temperature ($T<100$ K), while at higher temperature, the increase in the relative permittivity is very small. In addition to the temperature dependence of the relative permittivity on temperature, $\varepsilon_r$ is strongly affected by hydrostatic pressure. The application of hydrostatic pressure on STO leads to dramatic decrease in $\varepsilon_r$. A hydrostatic pressure of 2 kbar leads to a reduction in $\varepsilon_r$ by almost 60% at 4 K (see Fig.2.3). The application of hydrostatic pressure also extends the quantum paraelectric range to a higher temperature [9]. At ambient pressure, $\varepsilon_r$ shows Curie-Weiss behaviour down to 15 K, then it becomes constant. In addition to hydrostatic pressure, the relative permittivity of STO is also sensitive to electric field [10]. The relation is shown in Fig.2.4, where $\varepsilon_r$ decreases with increasing applied electric field.
2.2 Origin of charge carriers

Different hypotheses have been proposed to explain the origin of the charge carriers in LAO/STO heterostructures. The most important scenarios for the generation of charge carriers are described in the following.

2.2.1 Polar catastrophe

The most common hypothesis used to explain the conductivity in LAO/STO is called "polar catastrophe hypothesis", which was first proposed by Ohtomo el. al. [18] in 2004. It is based on the fact that the polarity (P) of LAO layers generates an electric field (E) within the LAO film, which causes an electrostatic potential (V) which builds up with increasing LAO film thickness, diverging eventually to infinity (see Fig.2.5).
The polarity in LAO generates an electric field within the LAO film, which causes the electrostatic potential to diverge to infinity with increasing film thickness, as seen in (a). Once the critical thickness (4 unit cells) of the sample is reached, the potential within the LAO becomes such high, that half of an elementary charge per unit cell is transferred from the LAO surface to the interface. This reconstruction causes the electric field to oscillate around zero (b) leading to a stable, non-diverging potential.

The polar discontinuity is not restricted to LAO/STO systems. It can also be found in other TMO heterostructures such as, NdGdO$_3$/STO, GdTiO$_3$/STO, LaTiO$_3$/STO and $\gamma$-Al$_2$O$_3$/STO, and likewise in semiconductors such as, GaAs/Si and GaAs/Ge [32]. There are several ways to avoid the effect of polar discontinuity at the interface. In GaAs/Si and GaAs/Ge, models propose that a roughening at the atomic scale, solves the problem [32]. In case of the LAO/STO interface, a 50% mixed surface termination (TiO$_2$ and SrO) can also avoid the polar catastrophe. In case of the LAO/TiO$_2$-STO interface, the buildup of the potential causes the energy of the LAO valence bands to increase. That increase in energy causes a charge transfer (half elementary charge per unit cell), from the surface of the LAO film to the interface, specifically to the Ti-3d$_{xy}$ orbitals closest to the Fermi energy $E_F$. This charge transfer makes the interface to be metallic. The expected sheet carrier concentration of the metallic interface equals to $n_s = 3.3 \times 10^{14} \text{cm}^{-2}$. Polar mismatch is also expected for (111) oriented LAO/STO heterostructure and hence should lead to interfacial doping as well [33].

### 2.2.2 Oxygen vacancies

The oxygen partial pressure, $p$(O$_2$), used during the deposition of oxide thin films, has direct impact on the oxygen content in the film, which crucially affects the film properties. It is well established that LAO/STO electrical properties are very sensitive to $p$(O$_2$) used during sample growth [18, 23]. Samples grown at relatively high oxygen pressure $p$(O$_2$) > 0.1 mbar, are usually...
insulating. In contrast, samples grown at lower $p(\text{O}_2)$ tend to be metallic. The dependence of conductivity of the samples on the oxygen partial pressure used during film deposition caused speculations that the origin of the charge carriers at the interface is due to the presence of oxygen vacancies ($V_\text{O}^-$) in the sample. The presence of $V_\text{O}^-$ results in a reduction of the Ti$^{4+}$ valence-state and hence likewise to the formation of non-localised charge carriers close to the interface.

2.2.3 Ionic intermixing

Another possible source for the generation of electrons at the interface between LAO and STO is atomic intermixing [34, 24]. During sample growth in PLD, atoms/ions with high kinetic energy (some tens of eV) are formed which may penetrate the substrate surface to some extent. This process usually leads to some atomic intermixing in the surface region resulting in a disorder site occupancy. In case of LAO/STO heterostructures, La$^{+3}$ ions may occupy Sr$^{+2}$ sites introducing one extra electron into the electron system. The valency of Ti may also reduce from +4 to +3. However it is doubtful whether the degree of intermixing is strong enough to be considered as a dominant source for free charge carriers in LAO/STO.

The exact contribution from each mechanism mentioned above to the metallic conductivity in LAO/STO is still under debate. It is unclear whether it is possible to create metallic LAO/STO heterostructure where only one of these mechanisms is exclusively present. One rather has to assume that more than one effect contributes to conductivity in LAO/STO. It seems that the polar discontinuity and the oxygen vacancies are the main sources of free electrons in LAO/STO. One can always try to minimize the contribution of oxygen vacancies and ionic intermixing to $n_s$ by carefully controlling the preparation and deposition conditions and analysing their impact on electronic transport properties.

2.3 Electronic properties

Crystal fields cause the degenerate Ti 3d orbitals to split into two $e_g$ and $t_{2g}$ states. Zhong et al. [11] used density-functional-theory (DFT) calculations to investigate the orbital reconstruction at the interface and found that the $t_{2g}$ orbitals undergoes further splitting caused by the constrained motion of electrons along the $z$ direction which is strongly influenced by the presence of the interface [35]. This splitting in energy at the $\Gamma$ point was also confirmed by X-ray absorption spectroscopy measurements [19] and by angular resolved photoemission (ARPES) [12], lowering energy of the 3d$_{xy}$ orbitals by about 200 meV compared to the degenerate 3d$_{yz}$.
2.3. Electronic properties

and 3d$_{xz}$. Fig.2.6 shows the theoretical calculation for the expected orbital reconstruction that occurs at the interface and the ARPES results. The lowering of the d$_{xy}$ orbitals energy is highest at the top layer of STO substrate. As for the layers buried deeper in the STO substrate, the splitting in the Ti-3d orbitals decreases for the layers buried deep in STO substrate, until eventually the orbitals become degenerate again as expected for bulk.

Figure 2.6: a) The DFT calculation for subband structure of LAO/STO heterostructures. The ARPES measurement results (b) of subbands structure of STO(001) surface. The drop in energy of d$_{xy}$ orbitals differs from one layer of STO to another. The highest drop in the d$_{xy}$ orbitals occurs at the top layer of the STO substrate, at the vicinity of the interface. As we dive deeper into the STO substrate, the splitting decreases per layer, until it reaches the value of zero, where the three 3dTi orbitals are degenerate again, as it is expected in bulk STO. Figures are taken from [11], [12] and [13].

The difference between 3d$_{xy}$ orbitals and 3d$_{xz}$ and 3d$_{yz}$ is that the former reside in the TiO$_2$ planes close to the interface, while the latter extend further into STO substrate in the direction perpendicular to the interface. This reconstruction causes the charge carriers to occupy the 3d$_{xy}$ orbitals first. As sheet carrier density increases and a certain charge carrier density is reached ($n_c = 1.7 \times 10^{13} \text{ cm}^{-2}$), a Lifschitz transition occurs, i.e., the Fermi-surface symmetry changes due to the additional occupation of 3d$_{xz}$ and 3d$_{yz}$ orbitals, leading to multi-band transport behaviour [15]. Conducting-tip AFM experiment show that the metallic interface has a thickness of about 7nm [36]. The system also exhibits Rashba splitting (splitting of the spin band in two-dimensional systems caused by the existence of spin-orbit coupling along side asymmetry of the orbitals in the direction perpendicular to the two-dimensional system). The splitting energy around the Γ point is only a few meV for 3d$_{xy}$ orbitals. The magnitude of this splitting can be modulated by the application of an external electric field [37].

The system also exhibits two-dimensional superconducting transition with a Berezinskii-
Kosterlitz-Thouless transition temperature $T_{BKT}$ of $\sim 250$ mK \cite{25}. This superconducting transition is found to be sensitive to the charge carrier density at the interface, which can be tuned by applying a back gate voltage to the sample \cite{14}. Tuning the charge carrier density near the superconducting transition temperature allows one to switch back and forth between the superconducting state and the normal state. Fig. 2.7 shows the critical temperature $T_{BKT}$ and the sheet resistance $R_S$ dependence on gate voltage. The superconducting region of the phase diagram is mapped by measuring $T_{BKT}$ under different values of applied gate voltages. The maximum value of $T_{BKT}$ amounts to about 315 mK, at gate voltage of about +120 V. A quantum critical point (QCP) can be deduced from the phase diagram using the scaling relation $T_{BKT} \propto (V - V_c)^{2/3}$ \cite{14}. The QCP occurs at $n_s = 1.5 \times 10^{-13}$ cm$^{-2}$.
Chapter 3

Experimental

3.1 Sample preparation

3.1.1 Substrate treatment

The fabrication of heterostructures and superlattices with full control over their composition and structure at an atomic level is one of the most exciting areas of condensed matter research. Using substrates with atomically flat surface is essential to obtain high-quality thin films. A well defined surface of the substrate enables researchers to fully control the structure of the thin film grown on top of it. One of the most used substrates for growing oxide thin films and heterostructures is SrTiO$_3$. The lattice constant and thermal expansion coefficient of STO is compatible with many interesting TMO’s such as high temperature superconducting cuprates. Together with the ability to produce STO in large size single crystals with high purity, made it a popular choice among thin film scientists.

As mentioned earlier, STO has a cubic perovskite structure with lattice constant of 3.905Å. Along the [001] direction, the STO structure can be viewed as alternating layers of a charge neutral SrO layer and a charge neutral TiO$_2$ layer. STO substrates with different orientations such as (110) and (111) are also widely used. The (110) oriented STO can be built up by alternating SrTiO$^{4+}$ and O$^{2-}_2$ layers while the (111) oriented STO structure can be constructed by alternating layers of SrO$^{3-}_3$ and Ti$^{4+}$ [31]. See Figs. 3.2 and 3.3.

Generally, the surface of as-received (001)-oriented STO substrates contains a mixture of both terminations SrO and TiO$_2$. By the use of chemical etching, the SrO layer can be removed from the surface. This process consists of chemical and thermal treatments [31, 30] and is routinely used to obtain a TiO$_2$ termination of (001) oriented STO substrates [38]. In this section,
the termination process of (001), (111), (110) oriented STO substrates is described in detail.

First, the substrate is soaked in ultrasonic bath of distilled water for 10 minutes. The water reacts with the strontium oxide creating strontium hydroxide (Sr(OH)$_2$) at the surface of the substrate.

$$\text{SrO} + \text{H}_2\text{O} \rightarrow \text{Sr(OH)}_2$$

After that, the substrate is etched for 30 seconds in a buffered hydrofluoric acid (BHF) solution which removes the strontium hydroxide (Sr(OH)$_2$) from the surface of the substrate. After that, the substrate is annealed under ambient oxygen flow and temperature of 950°C for 90 minutes. The heating and the cooling rates are set at 200°C per hour. Fig. 3.1 shows a schematic of the substrate cross section before and after the termination process.

![Figure 3.1: A sketch of a (001) oriented SrTiO$_3$ substrate before(a) and after(b) the chemical selective etching. In (a), the surface upper layer has a mixed termination of SrO and TiO$_2$. After the treatment process, the SrO is removed completely leaving TiO$_2$ terminated surface.](image)

In order to investigate the surface quality of the treated substrates, we studied the surface by atomic force microscopy (AFM) in contact mode. Fig. 3.2 (a) shows an AFM image of
3.1. Sample preparation

a treated (001) STO substrate surface (1µm × 1µm). The surface consists of atomically flat terraces with an average width of about 125nm and a step height of 0.39nm which compares to a single unit-cell height. Hence, the AFM measurements document the single-type termination of STO. For a mixed surface termination a step-height of \( a/2 \) should be present as well. The average width of the terraces is determined by the miscut angle \( \alpha \), by \( \tan(\alpha) = \frac{s}{w} \) where, \( s \) is step height and \( w \) is the terrace width accounting for \( \alpha = 0.5 \) for the shown STO substrate.

We can conclude that our (001) substrate is TiO\textsubscript{2}-terminated since the TiO\textsubscript{2} layer is stable and does not react with BHF. In order to directly determine the topmost atomic layer of the substrate, time-of-flight mass spectrometry (TOFMS), is usually performed. This technique is capable of high sensitive surface composition analysis. The results show that surfaces of substrates, treated with this method, are homogeneously TiO\textsubscript{2} terminated [39]. X-ray photo-electron spectroscopy (XPS) can also be used to investigate the substrate surface where XPS spectra are taken for Sr 3d, Ti 2p, and O 1s peaks under surface sensitive conditions [38].

![Figure 3.2: (a) AFM scan of (001) SrTiO\textsubscript{3} substrate after the TiO\textsubscript{2} termination process. The scan size is 1µm x 1µm. The height of the steps equals to 0.39nm. The terrace width is about 125nm. The crystal structure of a TiO\textsubscript{2}-terminated terrace is shown in (b).](image)

In the case of (111) substrates, it is difficult to obtain a well defined surface without atomic reconstruction because of the polar nature of its surface [38]. However, reports have shown that this treatment process is able to produce a well defined surface termination as well. Chemical etching and thermal annealing remove the SrO\textsubscript{3}^{-4} layer leaving a Ti-terminated surface.

For (110) substrates, the polarity of the surface is also an issue. Furthermore, the coexistence of Sr and Ti ions on the same plane adds more complexity to the system. Reports have shown that the treatment process results only in a "Ti-rich surface" [39]. The surface terraces are possibly not atomically flat and probably still contain some residual Sr. Therefore, a single-
3.1. Sample preparation

type termination of (110) oriented STO substrates is usually not possible.

Fig. 3.3 shows an AFM image of a treated (111) substrate and the structure of (111)-oriented STO(b) with a Ti-terminated surface (upper rows).

![AFM image and crystal structure](image)

Figure 3.3: (a) AFM image of (111) SrTiO$_3$ substrate surface after the BHF etching process. The image size is 500nm x 500nm. The height of the steps is 2.6 Å, and the terrace width is about 100nm. The step height indicates a single-type termination surface (Ti$^{4+}$). (b) The crystal structure of (111) oriented STO is also shown. The distance between the Ti$^{4+}$ and SrO$_3^{-4}$ layers, is $a/\sqrt{3}=2.25$ Å.

3.1.2 Film deposition

Pulsed laser deposition (PLD) is considered to be one of the most convenient and reliable methods to produce epitaxial multi-component oxide thin films with very low defect density. The development of this technique started in the 1960s but the major breakthrough came in 1987 when Dijkamp and Venkatesan [40] were able to successfully grow high-quality thin films of (the then newly discovered) high temperature superconductor YBa$_2$Cu$_3$O$_7$ using PLD technique. This achievement brought PLD to the spotlight and it became a mainstream technology.

In the PLD process, a solid target of the desired film material is exposed to focused high-energy laser pulses in a vacuum chamber, generating a high-temperature plasma which expands away from the target perpendicular to its surface. The material ejected from the target then condenses on a heated substrate located a few centimeters apart from the target. The fast and strong heating of the target surface by the intense laser pulse ensures that all target components evaporate at the same time irrespective of their partial binding energies and individual vapor pressures. This insures that the stoichiometry of the target can be retained to large extent in the deposited films. In fact, preserving the stoichiometry of the target material is the most cited advantage of PLD technique. The laser beam is directed using a set of mirrors to enter the vacuum chamber through a UV-transparent quartz window and is focused on the surface.
3.1. Sample preparation

of a rotating target using a lens. The vacuum chamber is kept at the desired oxygen pressure and at a constant oxygen flow by a set of valves and pumps. The process of the deposition is illustrated in Fig.3.4.

Figure 3.4: Schematic of the process of pulsed laser deposition. The valves and the pumps are not shown in the schematic.

At first glance, the PLD process seems to be very simple and straightforward but, in fact, many variables can affect the film forming process. The laser fluence, pulse frequency, distance between target and substrate, substrate temperature, residual gas pressure, and post deposition annealing time are some of the most important parameters that need to be optimized in order to produce high-quality thin films. In this study, many calibration runs are performed to optimize growth conditions for the production of heterostructures with metallic conductivity and to calibrate the growth rate of the film at these conditions. In the following the optimized growth parameters are described.
3.1. Sample preparation

A single crystal of LaAlO$_3$ is used as target to deposit LaAlO$_3$ thin films on TiO$_2$-terminated SrTiO$_3$ substrates forming LaAlO$_3$/SrTiO$_3$ heterostructures. The treated substrates are introduced to the deposition chamber where an atmosphere of $10^{-5}$ mbar oxygen is established. The substrates are then radiatively heated to 650°C. After that, a UV excimer laser with a wavelength of 248nm is used to ablate the target. The laser fluence is set at 1.1 J/cm$^2$ with frequency of 2Hz which results in a growth rate of about 0.11Å per laser pulse. After the deposition the film is cooled down to room temperature in $p$(O$_2$)=$10^{-5}$mbar. The growth rate is calculated by relating the thickness of a thin film to the number of pulses used to grow it. The thin film thickness can be measured by X-ray reflectivity. Fig. 3.5(a) shows an X-ray reflectivity measurement of a LAO film grown on STO (red curve) and the corresponding fit to the data (red curve) from which the film thickness and the roughness can be determined. A plot of film thickness versus number of pulses for a set of calibration runs is shown in (b). In our films the calculated growth rate is 0.11Å per pulse.
3.1. Sample preparation

3.1.3 Optimization of the oxygen partial pressure

The purpose of the optimization of the oxygen partial pressure $p(O_2)$ is to minimize the number of oxygen vacancies and their contribution to the conductivity in the LAO/STO heterostructures. The optimized $p(O_2)$ is the maximal $p(O_2)$ that can be used during sample growth while maintaining metallic behavior of the sheet resistance down to 4.2 K. We have grown several samples under different $p(O_2)$. Other deposition parameters, such as substrate temperature, laser energy and pulse frequency, were kept constant during the optimization process.

The oxygen partial pressure $p(O_2)$ was varied from $10^{-3}$mbar to $10^{-5}$mbar. The sheet resistance $R_S$ is plotted in Fig.4.1 versus $T$ for different $p(O_2)$. For $p(O_2) = 10^{-3}$mbar $R_S$ displays a minimum at $T_{MIN} = 47$K. Below this temperature, the sheet resistance increases with decreasing temperature down to 4.2 K. With decreasing $p(O_2)$ $T_{MIN}$ shifts down to lower $T$, and $R_S(4K)$ decreases considerably from 131kΩ/□ at $10^{-3}$mbar to 200Ω/□ at $10^{-5}$mbar. Only samples grown at $p(O_2)= 10^{-5}$mbar show complete metallic behaviour down to 4.2 K without displaying a minimum in $R_S$ vs $T$.

To further investigate the influence of oxygen vacancies in the STO substrate on the conductivity of LAO/STO heterostructures, an experiment was performed on a bare STO substrate without material deposition. The substrate was exposed to the same optimized conditions of the deposition process that were used to prepare LAO/STO heterostructures ($T_S = 650^\circ$C, $p(O_2) = 10^{-5}$mbar), however, without depositing LAO. The STO substrate handled under these conditions were highly insulating down to 4.2 K and did not show any conductivity. So we can rule out, that a significant amount of $V_{O^-}$ is formed during the PLD process, because of the low $p(O_2)$ used in the process. However, this does not mean, that $V_{O^-}$ are not present in the samples. The resulting atmosphere may also lead to the formation of reduced Al on top of the STO substrate during the LAO deposition which on the other side may lead to suck out of oxygen from the STO surface. Hence, formation of $V_{O^-}$ can happen. This process is well known in Al$_2$O$_3$ STO heterostructures [41] leading to a reduction of Ti and therefore metallic behaviour.
3.2 Structural properties

Figure 3.6: (001) oriented LAO/STO heterostructures prepared under optimized growth conditions (see text). The thickness of the LAO layer is 17.9 nm (40 monolayers). (a) $\theta/2\theta$ scan in the vicinity of the (002) reflection of STO. (b) Reciprocal space map in the vicinity of the (103) reflection of STO. The contour plot is on a logarithmic scale and shows the scattered intensity as a function of the scattering vector $\vec{q}$, expressed in non-integer Miller indices $h$ and $l$ of the STO substrate reflection referring to the azimuth reference (100) and the surface normal (001). The bright spots indicate the (103) peak positions of STO and LAO from which the in- and out-of-plane lattice parameters are deduced. The cross indicates (103) LAO plane position for fully relaxed (FR) LAO. (c) Spherical aberration-corrected cross-sectional HRTEM of the interfacial region of the LAO/STO heterostructures. Scale and directions are indicated.

Structural properties of the films were investigated by means of X-ray diffraction (XRD) and high resolution aberration-corrected transmission electron microscopy (HR-AC-TEM). XRD measurements were performed using a Bruker D8 diffractometer with CuK$\alpha$ radiation. All samples studied here were grown under optimized growth condition as discussed before in this
3.2. Structural properties

Chapter. Fig. 3.6 (a) shows a $\theta$-$\theta$ scan of a 17.9 nm film which displays a sharp reflection from the (002) lattice plane of the STO substrate at angle $2\theta = 46.5^\circ$ and to its right, the (002) lattice plane reflection of the pseudocubic/tetragonal LAO film. The high intensity of the LAO (002) Bragg peak, indicates the high quality of the film. The observation of the thickness fringes with high intensities on both sides of the LAO Bragg peak implies good and homogenous crystallinity alike.

The in-plane and out-of-plane lattice constants at room temperature were precisely measured by reciprocal space mapping (RSM). Fig. 3.6(b) displays a reciprocal space map of a LAO/STO sample as obtained by X-ray diffraction in the vicinity of the (103) lattice reflection. The LAO film is 40 monolayers thick. As the film thickness ($t$) increases, the out-of-plane lattice parameter $c$ increases from 3.72Å (fully strained state) and reaches the fully relaxed value of $c = 3.79\text{Å}$ for films thicker than 10 monolayers. The in-plane lattice parameter $a$ was found to be always completely strained for all films with thicknesses less than or equal to 40 monolayers i.e., $a = 3.9\text{Å}$ which is identical to the lattice parameter of STO.

The microstructural properties of these samples were investigated at the laboratory for electron microscopy in the group of Prof. Dagmar Gerthsen using HRTEM. Fig. 3.6 (c) shows a spherical aberration-corrected cross-section TEM image. The image shows a very high crystallinity of the LAO film and a nearly perfect interface between LAO and STO. No significant intermixing of ions (La/Sr or Ti/Al) at the interface was detected. The TEM analysis documents the atomically sharp single-type TiO$_2$-termination at the interface and high crystalline quality of the prepared LAO/STO heterostructures.
3.3 Dependence of the sheet resistance on the film thickness

The polar catastrophe hypothesis suggests that LaAlO$_3$ films need to have a film thickness of at least 4 monolayers to display a conductive interface. Heterostructures with a LAO layer less thicker are expected to be insulating. Hence, observing metallic conductivity in LAO/STO heterostructures with a LAO thickness less than 4 monolayers suggests that some other mechanisms responsible for metallic behaviour are at work.

For this purpose, LaAlO$_3$ films with different thicknesses were deposited on TiO$_2$ terminated STO substrates. After the deposition, the sheet resistance was measured at room temperature using the Van-der-Pauw technique. Fig. 3.7 shows the room temperature sheet resistance $R_s$ as a function of the LAO film thickness.

![Figure 3.7: Sheet resistance versus film thickness for different LaAlO$_3$ films deposited on TiO$_2$ terminated STO. The films with thickness below four unit cells are highly resistive. The error bars represent half unit cell, which is due to the roughness of the film.](image)

The sample with $t=2$ ML shows $R_s$ that exceeds our measurement limit. The sample
with $t=28$ ML has a $R_S$ of $3.76\,\text{MΩ/□}$. The sample with $t=5.64$ ML shows a drastic increase of conductivity with $R_S = 22\,\text{kΩ/□}$. For LAO films thicker than about $6$ ML, $R_S$ seems to saturate at about $11\,\text{kΩ/□}$. The most drastic change in conductivity seems to occur between $t=3$ ML and $t=6$ ML. The result is consistent with the critical thickness of LAO, i.e., $t_{cri}=4$ ML, suggested from the polar hypothesis [42].
3.4 Transport measurements under hydrostatic pressure

3.4.1 Pressure cell

The transport properties of the samples were investigated under hydrostatic pressure using a commercial two-layer clamp-type pressure cell from C+T Factory Co. (Tokyo, Japan). The cell has a total length of 70mm and an outer diameter of 25mm. The outer layer is made of CuBe and the inner layer of NiCrAl which enables to reach pressures to about 3 GPa. Inside the cell, a teflon capsule (with an inner diameter of 3.9mm and length of 17mm) is used to host the sample. A schematic of the pressure cell used in this work is shown in Fig.3.8.

![Figure 3.8: A schematic of the pressure cell used in this work. The samples and the pressure gauge (lead piece) were installed inside the teflon tube which was filled with pressure mediating medium. The wires from inside the teflon tube leave the pressure cell from the lower lock nut. The hole in the upper lock nut is used to apply pressure using a rod (not shown) made of tungsten carbide (WC).](image)

The sample (with dimensions $2.5 \times 2.5 \text{mm}^2$) is mounted with its surface normal oriented parallel to the capsule axis within $\pm 5^\circ$. After mounting the sample, the Pb-pressure gauge and the CuBe plug into the teflon capsule, the capsule is filled with a pressure-transmitting medium, namely Daphne 7373 oil (from the company Idemitsu, Tokyo, Japan). This oil ensures approximately hydrostatic pressure up to almost 2 GPa due to its high solidification pressure of
2.2 GPa at 298K [43]. The teflon tube is sealed with a copper beryllium (CuBe) plug. The plug has a small bore hole which allows feeding the wiring to the sample. After feeding all the wires through the copper plug, the bore hole is sealed with a two components glue of Stycast 2651\textsuperscript{TM} and catalyst 9\textsuperscript{TM} (Emerson\textsuperscript{TM} and Cumming\textsuperscript{TM}). The ratio of Stycast to catalyst by weight is 100 to 7. The glue is left for 24 hours to solidify. A hydraulic press is then used to pressurize the cell. With respect to the cross section of the teflon tube (3.9mm diameter), roughly 12kN is needed to achieve a pressure of about 1 GPa. Fig. 3.9 shows the CuBe plug after contacting the sample and a Pb piece serving as a pressure gauge.

![Image of CuBe plug and Pb piece](image_url)

Figure 3.9: The wiring through the copper beryllium plug after contacting the sample and the Pb piece. Both samples and Pb piece are installed inside the teflon tube which is filled with Dalphene 7373 oil as pressure-transmitting medium. The scale is indicated.

All transport measurements were carried out in an Oxford Cryostat. With that cryostat, we are able to measure down to 4.2 K and apply magnetic field up to 7 T. The standard sample holder delivered with the Cryostat was not suitable to host the pressure cell. For this reason, a new sample holder was designed in house. The holder has a tube-like cavity that hosts the pressure cell. After installing the pressure cell in the cavity, a lock nut is used to fix it in place and to prevent it from moving during the measurement. In order to accurately read temperatures and magnetic field during measurements, the temperature and hall sensors of this holder are
3.4. Transport measurements under hydrostatic pressure

installed as close to the pressure cell as possible. Test measurements were carried out to confirm
the accuracy of sensor readings of temperature. In these test measurements the pressure cell
was filled with Daphne 7373 oil but no pressure was applied and the superconducting transition
temperature of the lead piece was measured by an induction method. The measured $T_C$ of the
lead piece was 7.2 K which is consistent with literature [44]. This confirms that the readings from
the temperature sensor installed on the holder are accurate. Fig.3.10 shows the superconducting
transition of the Pb piece measured during cycling the temperature up and down in the vicinity
of $T_C$ with a sweep rate of 0.5K/min. The measurement was to account for the delay in
temperature acquirement of the cell. The difference between heating and cooling curves is
about 17mK. The $T_C$ is determined by taking the average of both curves.

Figure 3.10: $T_C$ measurements of the Pb piece installed in the pressure cell. The teflon tube was filled
with Daphne 7373 oil. The pressure cell was sealed before the measurement. The measurements are
taken at ambient pressure. For a temperature rate of 0.5K/min, the difference between heating (red
symbols) and cooling (black symbols) curves is about 17mK. The blue curve is the average of heating
and cooling curves. The unusual sign of the hysteresis might be because the heater position is closer to
the lead piece than the thermometer.

For measurements at ambient pressure, a special carrier was used instead of the pressure
3.4. Transport measurements under hydrostatic pressure

cell. Hence, the sample was mounted and contacted on a cylindrically shaped carrier made of copper with the exact dimensions of the pressure cell. The sample was mounted with its surface normal oriented parallel to the cylinder axis alike. A schematic and a photo of the bottom part of the sample holder, pressure cell and sample carrier are shown in Fig.3.11

Figure 3.11: (a) Schematic of the sample holder designed to host the pressure cell. The cell is inserted into the tube like a carrier. Hall and temperature sensors are also shown. The sample is located in the center of the teflon tube inside the pressure cell. (b) shows a photo of the pressure cell, the lower part of the sample holder and the sample carrier. The lower part is covered with Teflon tape to protect the wire coiled around the cylinder that works as a resistive heater. The locknut is also shown on the right of the photo, used to close the sample holder after installing either the pressure cell or the sample carrier.
3.4.2 Pressure determination

In order to determine the hydrostatic pressure inside the pressure cell, a piece of bulk Pb is used as a gauge. The large sensitivity of the superconducting transition temperature ($T_C$) of Pb to the applied pressure ($p$), i.e., $dT_C/dp = -0.365$ K/GPa \cite{44}, allows for very accurate determination of the pressure. The $T_C$ of the Pb is determined by inductive measurements, where a small coil is wrapped around the Pb piece to pick up the change of magnetic flux induced by its transition to the superconducting state. The coil and the Pb piece are inserted together with the sample inside the teflon capsule. Fig.3.12 shows $T_C$ of Pb at different applied pressures. The uncertainty of the temperature readings is within 17mK (see Fig.3.10), which translates into pressure accuracy of $\pm 0.046$ GPa.

![Figure 3.12: Superconducting transition of Pb at different hydrostatic pressure. The data shown here represent the average between the heating and the cooling curves for each pressure.](image)

3.4.3 Contact preparation

Contacts to the sample are made by an ultrasonic wire bonder using a 50$\mu$m thick Al wire. This technique commonly achieves good Ohmic contacts to the buried interface. Before bonding, the
film with the standard size of 5 x 5 x 1 mm$^3$ is cut down to 2.5 x 2.5 x 1 mm$^3$ to fit into the pressure cell. The bonds are placed at the corners of the square shaped sample surface to allow for Van-der-Pauw measurements, which will be discussed in detail in the next section. The bonds are reinforced using silver epoxy glue (EPO-TEK H20E) because of its ability to withstand high pressures. In addition, 0.1mm thick Cu wires are used to further enforce sample contacts (see Fig. 3.13). This reinforcement with thicker wires is also necessary to stabilize the sample position within the Teflon tube. The copper wires can be adjusted by bending to achieve the desired orientation and position of the sample. All the wires are fed through the CuBe plug and the lower CuBe lock nut and soldered to the connectors on the sample holder.

Figure 3.13: Sketch of the contacting process of the buried 2D electron system of a LAO/STO heterostructures using an ultrasonic wire-bonder. Silver epoxy glue and copper wires are used to further enforce the contacts.

To check Ohmic behaviour and stability of the sample contacts we carried out 2 point current-voltage ($I$-$V$) measurements. The measurements were done in the range from 1 $\mu$A to 100 $\mu$A. The results are shown in Fig. 3.14. The linear behavior of the $I$-$V$ curve indicates Ohmic behaviour in the used current range. A current of 10 $\mu$A was used for the transport measurements throughout this work.
3.4. Transport measurements under hydrostatic pressure

Figure 3.14: 2 point I-V-measurements performed on a LAO/STO sample after contact preparation (see text). The measurements were done at 4.2 K. The inset represents a linear fit to the data.

3.4.4 Van-der-Pauw technique

Transport properties of the samples are investigated by measuring the sheet resistance $R_S$, Hall voltage $V_H$ and magnetoresistance MR. All transport measurements are carried out in Van-der-Pauw (VdP) geometry [45]. The square-shaped (2.5x2.5mm$^2$) of the samples allows for nearly perfect measuring conditions if the contacts for the four-probe measurement are attached to the corners of the sample surface. The VdP technique requires a homogeneous conductivity of the sample with very small thickness. Using conformal mapping, Van-der-Pauw could show:

$$e^{-\pi d R_{12,34}/\rho} + e^{-\pi d R_{23,41}/\rho} = 1$$  \hspace{1cm} (3.1)

where $\rho$ is the resistivity, $d$ the film thickness and
3.4. Transport measurements under hydrostatic pressure

\[ R_{12,34} = \frac{U_{34}}{I_{12}} \]
\[ R_{23,41} = \frac{U_{41}}{I_{23}} \]  

(3.2)

In case of a square shape sample geometry, with the contacts 1-4 at the sample corners (see Fig. 3.15)

\[ R_{12,34} \approx R_{23,41} \approx R_{34,12} \approx R_{41,23} \]  

(3.3)

Figure 3.15: Schematic of the geometry used for VdP measurements in this work. The four yellow circles in the corners represent the places where the bonds are made. The sample has dimensions of 2.5mm x 2.5mm. The numbers refer to the contacts in VdP measurement.

The direction of the current is reversed for each measurement in order to remove the effect of the thermoelectric voltages generated at the contacts. These additional measurements increase the total number of measurements for each data point to eight. The average of all eight values gives us \( R_{\text{avg}} \).

This simplifies Eq. 3.1 to:

\[ \rho = \frac{\pi d}{\ln(2)} R_{\text{avg}} \]  

(3.4)

The sheet resistance \( R_S \) is then given by:

\[ R_S = \frac{\pi}{\ln(2)} R_{\text{avg}} \]  

(3.5)
3.4. Transport measurements under hydrostatic pressure

For the Hall measurements, the magnetic field $B$ is applied perpendicularly to the sample surface. The Hall coefficient is given by $R_H = \frac{d}{B} \Delta R_{13,24}$. The change in resistance $\Delta R$ is measured for positive (+) and negative (-) field direction. We likewise averaged $\Delta R$ by $(\Delta R_{13,24} + \Delta R_{24,13})$

$$V_H = \frac{V_{13} + V_{24}}{4}$$

(3.6)

Where

$V_{13} = V_{13}^+ - V_{13}^-$

$V_{24} = V_{24}^+ - V_{24}^-$

Fig.3.16 shows the negative Hall voltage for a LAO/STO sample under hydrostatic pressure at three different temperatures. The Hall voltage is linear versus the magnetic field up to at least 7 T. This behaviour indicates one type of negative charge carriers. In this case, the sheet carrier density and the Hall mobility can be deduced by: $n_s = \frac{1}{\mu_H}$ and $\mu_H = \frac{1}{n_s e}$. The non-monotonic $T$-dependence of $V_H$ arises from the non-monotonic $R_s(T)$ dependence (see Fig. 5.1)
Figure 3.16: The negative Hall voltage versus the magnetic field for LAO/STO. The measurements are taken at \( p = 1.64 \) GPa at three different temperatures, \( T = 4.2 \) K, 48 K and 150 K. The linear behaviour indicates one-type-carrier-transport.
Chapter 4

Electronic transport properties of LaAlO$_3$/SrTiO$_3$ heterostructures at ambient pressure

4.1 Transport properties of LaAlO$_3$/SrTiO$_3$ grown under different oxygen partial pressure $p$(O$_2$)

Transport properties of LAO/STO heterostructure are very sensitive to the oxygen partial pressure during deposition. Generally, the conductivity increases with decreasing oxygen partial pressure [46]. The reason for that is probably that oxygen vacancies in the STO substrate are formed during the deposition process of LAO at low $p$(O$_2$) [23]. Oxygen vacancies are a source of electrons in these heterostructures, and can give rise to electrical conductivity. In this chapter, we represent and discuss the sheet resistance ($R_S$), sheet carrier concentration ($n_S$) and Hall mobility ($\mu_H$) as a function of the oxygen partial pressure used during sample growth.

4.1.1 Sheet Resistance ($R_S$)

In Fig.4.1, we have shown the sheet resistance $R_S$ versus $T$ for different $p$(O$_2$) ranging from $10^{-8}$mbar upto $10^{-3}$mbar.
4.1. Transport properties of LaAlO$_3$/SrTiO$_3$ grown under different oxygen partial pressure $p$(O$_2$)

The sample grown at $p$(O$_2$)=$10^{-3}$mbar shows the highest sheet resistance over the whole temperature range. At 4.2 K, the sheet resistance of this sample is about three orders of magnitude higher than the sheet resistance of the sample grown at $p$(O$_2$)=$10^{-5}$mbar. For $p$(O$_2$)=$10^{-4}$mbar and $2.5\times10^{-5}$mbar, $R_S$ amounts to 28kΩ/□ and 17kΩ/□ at room temperature and decreases to 6kΩ/□ and 1.9kΩ/□ at 4.2 K, respectively. Heterostructures grown at $p$(O$_2$)=$10^{-5}$mbar display the lowest $R_S$, it drops from 8800Ω/□ at room temperature to 203Ω/□ at 4.2 K which gives a ratio of $\frac{R_S(300K)}{R_S(4.2K)}=43.35$. At higher temperatures ($T > 200$ K), $R_S$ of samples grown at $p$(O$_2$) <$10^{-3}$mbar shows nearly a $T^2$ dependence. The green lines in Fig.4.1 represent power-law fits to the sheet resistance for $T > 120$ K. The values of the exponents are 2.1 and 2.0 for $p$(O$_2$) = $2.5\times10^{-5}$mbar and $p$(O$_2$) = $10^{-5}$mbar, respectively. Van der Marel et. al. [47] argued that electron-phonon interaction leads to a renormalization of the bare electron mass, density of states and plasma frequency by a factor of two, which enhances the electron-electron
scattering, leading to a $T^2$ behaviour up to 120 K. However, for $T > 100$ K the electron-phonon scattering increasingly contributes to the resistance leading to deviations from the exponent 2.

In addition, samples grown at $p(O_2) > 10^{-5}$ mbar exhibit a change to nonmetallic behaviour, i.e. $\frac{\partial R_S}{\partial T} < 0$. The minimum in $R_S$ versus $T$ at $T=T_{\text{min}}$ changes from sample to sample, and seems not to be dependent systematically on $p(O_2)$. With increasing $p(O_2)$, $T_{\text{min}}$ first increases from about 30 K to 50 K. However, for $p(O_2)=10^{-3}$ mbar, $T_{\text{min}}$ decreases again to 30 K.

Samples grown at high $p(O_2) = 10^{-3}$ mbar show a small dip in the sheet resistance ($R_S$) at $T=87$ K and $T=186$ K. S. Seri et. al. [48] argued that the increase in conductivity at these temperatures is due to the recovery of trapped charge carriers by thermal activation. The trapping of charge carriers, at low temperatures, might be caused by oxygen vacancies [49], by lattice dislocations found in LAO/STO close to the interface [50] or by twin boundaries which may form due to the structural phase transition at $T \approx 105$ K in STO [51]. Heating the sample close to or above that temperature likely activates charge carriers leading to a lowering of $R_S$.

### 4.1.2 Sheet carrier density and Hall mobility

Hall measurements are performed on samples grown at $p(O_2) = 10^{-5}$ mbar (Sample A) and $p(O_2) = 2.5 \times 10^{-5}$ mbar (Sample B) in the temperature range from 4.2 to 300 K. The sheet carrier concentration ($n_s$) and the Hall mobility ($\mu_H$) are deduced as explained before. The results are shown in Fig. 4.2 and Fig. 4.3.

At room temperature, sample A and sample B have a sheet carrier density of $n_s = 1.26 \times 10^{14}$ cm$^{-2}$ and $7.3 \times 10^{13}$ cm$^{-2}$, respectively. The slight difference in $n_s$ is likely due to the formation of charge carriers as a result of higher content of oxygen vacancies ($V_{\text{O}}$). $n_s$ for both samples decreases with decreasing temperature, as shown in Fig. 4.2. The charge-carrier-freeze-out effect might be caused by $V_{\text{O}}$ sites, which may act as charge trapping potentials [49]. It is well known that thermal activation of charge carriers is present in LAO/STO heterostructures [48]. The theoretical value of $n_s$ generated at the interface, according to the polar catastrophe hypothesis, is $3.3 \times 10^{14}$ cm$^{-2}$, i.e., $e/2$ per surface unit cell. Our results show that $n_s$ of sample A at room temperature is about one third of that value and about one order of magnitude less at 4.2 K. Nevertheless, these observed values of $n_s$ are in good agreement with values reported by other groups [52, 15]. For both samples, $n_s \approx 1.7 \times 10^{13}$ cm$^{-2}$ at 4.2 K, which compares well to the critical sheet carrier density ($n_c$) in LAO/STO [15], at which the $d_{xy}$ orbitals are nearly completely filled, and $d_{xz}$ and $d_{yz}$ orbitals begin to be occupied, resulting in a one-type
4.1. Transport properties of LaAlO$_3$/SrTiO$_3$ grown under different oxygen partial pressure $p(O_2)$

charge-carrier transport for $n_s \leq n_c$.

Figure 4.2: Charge carrier density ($n_s$) of two LaAlO$_3$/SrTiO$_3$ heterostructures grown at different oxygen partial pressures as a function of $T$. The lower line in the figure refers to the value of the critical density reported [15], at which a Lifshitz transition is expected to occur. The upper line is the theoretical value of the carrier density expected from the polar catastrophe hypothesis, i.e., $n_s = e/2$ per surface unit cell.

The mobility of the two samples is also deduced from the Hall measurements. The results are shown in Fig.4.3. At room temperature, both samples show the same mobility, which equals to 4 cm$^2$V$^{-1}$s$^{-1}$. At higher temperatures ($T$ > 50 K), the mobility of both samples displays the inverse $T$-dependence compared to $R_S$ which is proportional to $T^{-2.2}$. At lower temperatures ($T$ < 50 K), sample A shows a higher mobility (1230 cm$^2$V$^{-1}$s$^{-1}$) than sample B displaying a mobility of 196 cm$^2$V$^{-1}$s$^{-1}$. The increase in the mobility with decreasing temperature is due to the decrease of electron phonon scattering. However, below 50 K, the $T$-dependence of $\mu_H$ no longer follows the $T^{-2.2}$ behaviour, and becomes constant. The change in the $T$-dependence is related to the domination of impurity scattering, which is expected to be $T$-independent, resulting in $\mu_H(T) \approx$ constant at low $T$. Obviously, for $T$ > 50 K, electron-phonon scattering is the dominant scattering source, while for $T$ < 50 K impurity scattering becomes the dominant source.
4.1. Transport properties of LaAlO$_3$/SrTiO$_3$ grown under different oxygen partial pressure $p$(O$_2$)

Figure 4.3: Hall mobility versus $T$ of LaAlO$_3$/SrTiO$_3$ heterostructures grown at different $p$(O$_2$). The Hall mobility of both samples decreases with increasing temperature, following a $T^{-2.2}$ dependence for $T > 50$ K, where the dominant source for charge carrier scattering is electron-phonon scattering. The yellow line represents a linear fit to the data for $T>50$ K.
Chapter 5

Electronic transport properties of LaAlO$_3$/SrTiO$_3$ heterostructures under hydrostatic pressure

In this chapter, the electronic transport of LAO/STO heterostructures is studied as a function of the hydrostatic pressure applied to the sample. The measurements were carried out from ambient pressure to $p=1.64$ GPa. Electronic transport was measured as a function of temperature and magnetic field. The samples used in the study were grown under optimized deposition conditions and $p$(O$_2$)=$10^{-5}$mbar. Here we focus on the results.

5.1 Sheet resistance ($R_s$)

The sheet resistance for LAO/STO is measured in the temperature range of 4-300 K, under different values of hydrostatic pressure. The results of the measurements are shown in Fig.5.1.

For temperatures $T > 100$ K, the $T$ dependence of $R_s$ seems to depend only minor on hydrostatic pressure displaying roughly a $T^2$ dependence. The $T$-dependence in that temperature range is attributed to electron-phonon scattering as discussed in the previous chapter. For temperatures below 100 K, the effect of the pressure on the $R_S$ is more pronounced. $R_S$ at ambient pressure shows metallic behaviour down to 4.2 K. Under hydrostatic pressure, a minimum in $R_S$ at($T_{min}$) develops, followed by non-metallic behaviour down to 4.2 K. $T_{min}$ systematically increases with increasing hydrostatic pressure, from $T_{min}$=34K for $p=0.36$ GPa, to $T_{min}$=55K for $p=1.64$ GPa. $R_S$ at $T=4.2$ K increases with increasing hydrostatic pressure as well. $R_S$ amounts to 548Ω/□ at ambient pressure and increases to 1.4kΩ/□ at $p=0.36$ GPa,
5.1. Sheet resistance ($R_s$)

Figure 5.1: The sheet resistance $R_s$ of LAO/STO sample grown under standard conditions as mentioned in chapter 2, under ambient and hydrostatic pressure. $R_s$ at ambient pressure shows metallic behaviour down to 4.2 K. Under hydrostatic pressure, a minimum in $R_s$ develops. Below that, the sheet resistance starts to increase with decreasing temperature.

$R_s = 1.6 k\Omega/\square$ at $p = 0.63$ GPa and finally seems to saturate at the same $R_s$ for higher pressures.

In the temperature range 10-100 K, $R_s$ is dominated by electron-electron scattering enhanced by electron-phonon interaction ($R_{el-\text{el}}$) and impurity scattering $R_{imp}$. As demonstrated by van der Marel et. al. [47], $R_{el-\text{ph}}$ in STO is well described by a quadratic $T$ dependence, i.e., $R_{el-\text{ph}} \propto T^2$ in that temperature range as explained in section 1 in chapter 4. In order to model the impurity scattering, we assume the existence of localized charged impurities that can result, for example, from trapped charge carriers. Oxygen vacancies in STO are known to act as traps for carriers at low $T$. These trapped charge carriers can be thermally activated when the temperature is close to or above the activation temperature $T_A$ [48, 49]. Once charged, these impurities start to act as scattering centers for free electrons reducing their mobility and hence,
5.1. Sheet resistance ($R_s$)

increase $R_s$. The number of charged impurities as a function of temperature can be described by an Arrhenius law:

$$N(T) = N_0 \left[1 - \exp \left(-\frac{T_A}{T}\right)\right]$$

(5.1)

$T_A$ is the activation temperature and $N_0$ the total number of charged impurities.

The equation above indicates that the number of charged impurities decreases exponentially with increasing temperature, leading to a reduction of impurity scattering with increasing $T$. This reduction leads to an increase in carrier mobility, which again reduces $R_s$ with increasing $T$.

In addition to the number of charged impurities, $N(T)$, the magnitude of the scattering potential ($V$) of the impurities must be also considered. Because of the high dielectric permittivity $\varepsilon$ of STO at low $T$, screening of scattering potentials, i.e. $V \propto \varepsilon^{-1}$ may become very effective below 50 K strongly affecting impurity scattering.

As discussed in chapter 2.1, the dielectric permittivity of STO is very sensitive to hydrostatic pressure at low temperatures, decreasing significantly with increasing hydrostatic pressure [9]. The decrease of $\varepsilon$ with increasing $p$ leads to a decrease in screening of the impurity potential. At $p=\text{constant}$, $\varepsilon$ decreases with increasing $T$, leading to an additional reduction in the screening behavior alike. Therefore, the temperature dependence of $\varepsilon(T)$ must also be considered in order to model $R_{\text{imp}}$ correctly. As discussed in chapter 2.1, $\varepsilon(T)$ is well described by the Barrett formula [53]:

$$\varepsilon(T) = C \left(\frac{T_1}{2} \coth \left(\frac{T_1}{2T}\right) - T_0\right)^{-1}$$

(5.2)

$C$ is the Curie constant, $T_1$ is the temperature at which the quantum properties of the sample become important and $T_0$ corresponds to the classical mean-field $T_c$. To is reported to be [9] strongly sensitive to hydrostatic pressure decreasing from 35K at ambient pressure to zero for $p > 0.16$ GPa.
According to Fermi’s Golden Rule, impurity scattering is proportional to the number of impurities and the square of the scattering potential of the impurities:

\[ R_{\text{imp}} = N \times V^2 \]  

(5.3)

We assume that the impurity scattering potentials \( V_o \) is screened by \( \varepsilon(T) \), i.e. \( V = V_o/\varepsilon(T) \). The temperature dependence of \( R_{\text{imp}}(T) \) therefore results in:

\[ R_{\text{imp}} = A \left[ 1 - \exp \left( -\frac{T_A}{T} \right) \right] \left[ \frac{T_1}{2} \coth \left( \frac{T_1}{2T} \right) - T_0 \right]^2 \]  

(5.4)

where the constant \( A = \frac{N_o}{\varepsilon} \).

According to Matthiessen’s rule, which can be applied in case of isotropic independent sources of electron scattering, the total resistance is given by the sum of all the partial individual resistance which leads to:

\[ R_s = R_{\text{imp}} + R_{\text{el-ph}} \]  

(5.5)

and

\[ R_s(T) = A \left[ 1 - \exp \left( -\frac{T_A}{T} \right) \right] \left[ \frac{T_1}{2} \coth \left( \frac{T_1}{2T} \right) - T_0 \right]^2 + B \times T^2 \]  

(5.6)

In order to deduce the parameters \( A, T_A, T_1 \) and \( B \), we fitted Eq.5.6 to the data \( R_s(T, p) \) shown in Fig.5.1 in the temperature range from 10-100 K. The fitting results are shown in Fig.5.2.
5.1. Sheet resistance ($R_s$)

to Fig.5.6. The partial contributions to $R_S$, $R_{imp}$ and $R_{el-ph}$, are plotted herein for better comparison over the complete measured temperature range. The $T$-dependence for $T>100$ K of $R_{imp}$ and $R_{el-ph}$ were assumed to be the same as given in Eq.5.6.

Figure 5.2: Experimental data $R_s$ (red symbols) of the LAO/STO sample under ambient pressure. A fit (black symbols) to the data and the deduced partial contributions to $R_s$, i.e., $R_{imp}$ (blue symbols) and $R_{el-el}$ (orange symbols) are displayed alike. The $T$ dependence of $R_{imp}$ and $R_{el-el}$ was assumed to be the same over the complete temperature range.
Figure 5.3: Experimental data $R_s$ (red symbols) of the LAO/STO sample under $p=0.36$ GPa. A fit (black symbols) to the data and the deduced partial contributions to $R_s$, i.e., $R_{\text{imp}}$ (blue symbols) and $R_{\text{el-el}}$ (orange symbols) are displayed alike. The $T$ dependence of $R_{\text{imp}}$ and $R_{\text{el-el}}$ was assumed to be the same over the complete temperature range.
5.1. Sheet resistance \( (R_s) \)

Figure 5.4: Experimental data \( R_s \) (red symbols) of the LAO/STO sample under \( p=0.63 \) GPa. A fit (black symbols) to the data and the deduced partial contributions to \( R_s \), i.e., \( R_{\text{imp}} \) (blue symbols) and \( R_{\text{el-el}} \) (orange symbols) are displayed alike. The \( T \) dependence of \( R_{\text{imp}} \) and \( R_{\text{el-el}} \) was assumed to be the same over the complete temperature range.
Figure 5.5: Experimental data $R_s$ (red symbols) of the LAO/STO sample under $p=1.41$ GPa. A fit (black symbols) to the data and the deduced partial contributions to $R_s$, i.e., $R_{\text{imp}}$ (blue symbols) and $R_{\text{el-el}}$ (orange symbols) are displayed alike. The $T$ dependence of $R_{\text{imp}}$ and $R_{\text{el-el}}$ was assumed to be the same over the complete temperature range.
5.1. Sheet resistance ($R_s$)

Figure 5.6: Experimental data $R_s$ (red symbols) of the LAO/STO sample under $p=1.64$ GPa. A fit (black symbols) to the data and the deduced partial contributions to $R_s$, i.e., $R_{imp}$ (blue symbols) and $R_{el-el}$ (orange symbols) are displayed alike. The $T$ dependence of $R_{imp}$ and $R_{el-el}$ was assumed to be the same over the complete temperature range.
5.1. Sheet resistance ($R_s$)

The parameters obtained from the fit of equation 5.6 to the data of $R_s(T,p)$ are given in Table 1.

<table>
<thead>
<tr>
<th>$p$(GPa)</th>
<th>$T_A$(K)</th>
<th>$A$(Ω/K²)</th>
<th>$T_1$(K)</th>
<th>$B$(Ω/K²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>44</td>
<td>0.26</td>
<td>38.2</td>
<td>0.07</td>
</tr>
<tr>
<td>0.36</td>
<td>44</td>
<td>0.23</td>
<td>149.5</td>
<td>0.07</td>
</tr>
<tr>
<td>0.63</td>
<td>49.6</td>
<td>0.18</td>
<td>189</td>
<td>0.07</td>
</tr>
<tr>
<td>1.41</td>
<td>67.8</td>
<td>0.1</td>
<td>245</td>
<td>0.09</td>
</tr>
<tr>
<td>1.64</td>
<td>75.2</td>
<td>0.07</td>
<td>299</td>
<td>0.11</td>
</tr>
</tbody>
</table>

At ambient pressure, $T_A$ amounts to 44 K. Similar activation temperatures for charge carriers were also reported. [48, 49]. $T_A$ increases with pressure to 75K at $p=1.64$ GPa. $T_1$, the transition to the quantum paraelectric state, increases from 38.2 K at ambient pressure to 299 K at $p=1.64$ GPa. An increase of $T_1$ with increasing $p$ was also reported [9]. The parameter $A$ steadily decreases with increasing pressure which is likely related to a decrease of the Curie-constant under pressure. The parameter $B$, related to the strength of electron-phonon interaction increases only slightly with increasing $p$.

Comparing $R_{imp}$ and $R_{el-ph}$ for $p$ larger than ambient pressure, a striking feature is that in contrast to the monotonic increase of $R_{el-ph}$ with increasing $T$, $R_{imp}$ displays a pronounced minimum in the temperature range from 50-100 K.

That minimum is obviously the reason for the minimum in $R_s(T)$ and caused by the different temperature dependence on $N(T)$ and $V(T)$ determining $R_{imp}$ (see Eq. 5.3.) $N(T)$ decreases with increasing $T$, whereas $V(T)$ increases. To document the different $T$ dependence on $N(T)$ and $V(T)^2$ determining the $T$ dependence of $R_{imp}(T)$, we have plotted in Fig.5.7 the normalized number of impurities, $N(T)/N_o=N_N(T)$, and the squared inverse of the normalized permittivity ($\varepsilon(T)/\varepsilon(4.2 \text{ K})$)$^{-2} = \varepsilon_N^{-2}$ as a function of $T$ for $p=0.36$ GPa and $p=1.46$ GPa.

The temperature that marks the onset of a significant $T$ dependence of $N_N(T)$ and $\varepsilon_N^{-2}(T)$ is defined by $T^*$ and $T_q$, respectively. In case of $T_q>T^*$, the $T$-dependence of $R_{imp}$ is controlled by $N_N(T)$ for $T<T_q$, i.e., decreases above $T^*$ with increasing $T$. However, due to the stronger increase of $\varepsilon_N^{-2}$ with increasing $T$ above $T_q$, $R_{imp}(T)$ is controlled by the $T$-dependence of $\varepsilon_N^{-2}$, i.e., increases with increasing $T$, therefore leading to a minimum in $R_{imp}$. 
5.1. Sheet resistance ($R_s$)

$T^*$ and $T_q$ obviously depend very sensitively on $p$ versus the parameters $T_A$ and $T_1$ (see Table.1). With increasing $p$, $T^*$ and $T_q$ shift to higher temperature leading to a shift of the minimum of $R_{imp}(T)$ and hence $R_S(T)$, alike.

![Graph showing $N_N(T)$ and $\varepsilon_N(T)^{-2}$ as a function of $T$ for $p=0.36$ GPa (black symbols) and $p=1.46$ GPa (red symbols). For $T>T^*$ and $T>T_q$, $N_N(T)$ and $\varepsilon_N(T)^{-2}$ become $T$-dependent, respectively. Both $T^*$ and $T_q$ shown for $p=1.46$ GPa increase with increasing hydrostatic pressure.](image)

$\varepsilon_N^{-2}$ not only increases with $T$, but also with increasing $p$. The quite strong $p$ dependence of $\varepsilon(T,p)$ enters the permittivity via the pressure sensitivity of the quantum paraelectric state. As displayed in Table.1, the transition temperature $T_1$ to the quantum paraelectric state shows strong $p$-dependence affecting $\varepsilon$ directly, i.e. $\varepsilon \propto T_1^{-1}$.

In order to demonstrate the $p$ dependence of $\varepsilon_N^{-2}(T)$ we have plotted in Fig.5.8 $(C/\varepsilon(T))^2$ versus $T$ for different hydrostatic pressure. The curves were generated using the parameter $T_1$ as deduced from experiment (shown in Tab.1). The $p$- and $T$-dependence is well comparable to that reported in [9] for bulk STO. $\varepsilon(p,T)^{-2}$ increases with increasing $p$, where the sensitivity to
5.1. Sheet resistance ($R_s$)

$p$, $\frac{\partial \varepsilon^{-2}}{\partial p}$, seems to decrease with increasing $p$.

Coming back to $R_s(T, p)$, in the low-temperature range $T<10$ K, the extrapolation of the fitting curves indicates nearly $T$-independent behaviour, where $R_s(T, p)$ shows a small increase with decreasing $T$, which is obviously not described by Eq. 5.6. This will be discussed in more detail in chapter 5.3.

Figure 5.8: $C^2/\varepsilon (T)^2$ versus temperature at different hydrostatic pressure. The curves are generated using the parameter $T_1$ as deduced from experiment shown in Table 1. The $p$- and $T$-dependence is well comparable to that reported for bulk STO [9].
Despite the increase of the sheet resistance with increasing hydrostatic pressure at lower temperatures, the sheet carrier density ($n_s$) increases by a factor of three. Fig.5.9 shows that the sheet carrier concentration as a function of temperature measured under different hydrostatic pressure. In the intermediate temperature range $n_s(T)$ dips around 60 K. The increases of $n_s$ may be explained by structural changes of STO or LAO under hydrostatic pressure. The bulk modulus for both LAO and STO is nearly the same for $p < 10$ GPa [54, 55], which means that the pressure close to the interface should also be hydrostatic. For $p < 2$ GPa, the STO does not exhibit any symmetry change [55]. Therefore, we do not expect any significant change in the energetic succession of t$_{2g}$ sub-bands, which means t$_{2g}$ are still the orbitals with the lowest energy and the first available to be occupied. Applying hydrostatic pressure on bulk STO shrinks its unit-cell volume by about 1%, primarily by equal bond-length reduction [56]. Therefore, the distance between atoms is reduced, increasing the crystal fields. Hence, the splitting between Ti 3d e$_g$ and t$_{2g}$ states increases and shifts the t$_{2g}$ states downward in energy easily by a few meV [57], which may result in an enhanced thermal population by donor states. Biaxial compression of 1-2% in LAO/STO likewise leads to a significant increase of $n_s$ [58]. In addition, structural changes in LAO have to be taken into account as well. The polar discontinuity at the
interface is accommodated through an intricate combination of ionic relaxation and electronic reconstruction which partially may compensate excess charge and thus lead to \( n_s < n_p \) \[59\].

On the other hand, hydrostatic pressure may diminish the resulting buckling leading to an increase of \( n_s \). For \( T < 20 \) K, our sheet carrier density is comparable with the sheet critical density \( n_c \) reported in \[15\], where a Lifshitz transition between \( d_{xy} \) and \( d_{xz}, d_{yz} \) is expected at \( n_c = 1.7 \times 10^{13} \text{cm}^{-2} \). As temperature increases, more carriers are activated and start to occupy \( d_{xz} \) and \( d_{yz} \) orbitals leading to a two-type-carrier transport for \( n_s > n_c \). The one-type-carrier transport is expected only when \( n_s < n_c \), where only \( d_{xy} \) orbitals are occupied. In Fig.5.10, we plot \( \ln(n_s) \) versus \( 1/T \) for temperatures \( > 50 \) K at ambient pressure. The red line represents the linear fitting for the data. The relation between charge carrier density and the activation energy can be expressed by \( n_s(T) = e^{-\frac{E_A}{k_B T}} \), which leads to an activation energy of 5.8meV.

In contrast to the increase of the sheet carrier density with increasing pressure, measurements show that the Hall mobility decreases with increasing \( p \) and decreases with increasing \( T \). The results of the measurements are shown in Fig.5.11.
5.1. Sheet resistance ($R_s$)

The Hall mobility ($\mu_H$) as a function of $T$ for LAO/STO sample grown at $p(O_2)=10^{-5}$ mbar for different hydrostatic pressure. For $T > 110$ K the electron-phonon scattering is the dominant source of scattering.

![Figure 5.11:](image)

The Hall mobility decreases with increasing the hydrostatic pressure at the low temperature range ($T < 60$ K), from $700$ cm$^2$V$^{-1}$s$^{-1}$ at ambient pressure, to $60$ cm$^2$V$^{-1}$s$^{-1}$ at $p=1.64$ GPa and displays a $T$-independent behavior at a much lower level than observed at ambient pressure.

The sheet resistance relates to the Hall mobility ($\mu_H$) and carrier concentration ($n_s$) as $R_s \propto (n_s \mu_H)^{-1}$. The increase in $R_s$ below $T_{min}$ suggests that $\mu_H$ has much more impact in determining the conductivity in the LAO/STO than $n_s$. As shown earlier, $n_s$ actually increases with increasing $p$, however, it seems that this increase is completely dominated by the scattering of the carriers caused by the charged impurities. The strong decrease of $\mu_H$ with increasing $p$ can be explained by the strong decrease of the static dielectric permittivity $\varepsilon$ of STO. $\mu_H$ scales inversely with the scattering rate which, according to Fermi’s golden rule for impurity scattering, is proportional to $V^2$ as discussed before. Hence in the low-temperature regime $\mu_H \propto \varepsilon^2$ is expected. Fig.5.12 shows $\mu_{Hn} = \mu_H(p)/\mu_H(0)$ and $\varepsilon_n^2 = |\varepsilon(p)/\varepsilon(0)|^2$ versus $p$ at 4.2 K. Both curves show similar pressure dependence, which suggests direct impact of the dielectric permittivity on Hall mobility.

Magnetoresistance (MR) measurements were performed on a LAO/STO heterostructure grown at $p$(O$_2$) = 10$^{-5}$mbar, $MR(H) = [R_s(H) - R_s(0)]/R_s(0)$ for LAO/STO under ambient and a pres-

sure of 1.64 GPa, at $T = 4.2$ K, 48 K and 150 K where $R_s(H)$ is the sheet resistance as a function of magnetic field and $R_s(0)$ is the sheet resistance at $\mu_0H=0$. MR versus $\mu_0H$ curves for LAO/STO are shown in Fig.5.13.

![Figure 5.13: Magnetoresistance measurements versus $\mu_0H$ for the LAO/STO sample grown at $p(O_2) = 10^{-5}$ mbar. The measurements are done under ambient pressure and at $T = 4.2$ K, 48 K and 150 K. MR is positive for the three temperatures. The maximum increase in the sheet resistance is observed at $T = 4.2$ K. MR decreases as $T$ increases for the same field.](image)

At ambient pressure, MR is always positive for the three temperatures and scales with $1/R_s(0)$ of the sample, i.e., MR strongly increases with decreasing $R_s$. The largest increase in the sheet resistance is observed at temperature of $T = 4.2$ K, which is equal to 20% at $\mu_0H = 7T$. The lowest MR is observed at $T = 150$ K, which is about 1%. The sheet resistance for the sample at $T = 4.2$ K, 48 K and 150 K are 90Ω/□, 530Ω/□ and 2.73kΩ/□ respectively. The observed field dependence seems to be typical for LAO/STO [15]. The field dependence of MR at ambient pressure displays a quadratic dependence on $H$ up to 4T at 4.2 K followed by

negative curvature. With increasing $H$ and $T$ MR displays contributions linear in $H$ [61]. MR at low fields is obviously dominated by classical defect scattering following Kohler’s rule, i.e. $\text{MR} \propto [H/R(0)]^2$. The increases in magnetic field increases the distance the electron travels due to the helical path of the electron moving in a magnetic field, leading to an increases in scattering rate, leading to an increase in $R_s$ with increasing $H$.

MR under hydrostatic pressure of $p=1.64$ GPa was also measured for the same temperatures. The results are shown in Fig.5.14

![Figure 5.14: Magnetoresistance (MR) versus $\mu_0 H$ for the LAO/STO heterostructures. The measurements were done at temperature of 4.2 K, 48 K and 150 K under hydrostatic pressure of $p=1.64$ GPa.](image)

At $p=1.64$ GPa, MR is also positive and is one order of magnitude less than ambient pressure (see Fig.5.14). The drop in MR is expected as $R_s$ is increasing with increasing hydrostatic pressure. The MR at temperature of 150 K and 48 K show the effect of the classical scattering behaviour, as in the ambient pressure measurements. However, at $T=4.2$ K MR is reduced
Magnetoresistance measurements on LaAlO$_3$/SrTiO$_3$. strongly compared to the ambient pressure measurement at the same temperature possibly due to the large increase of the sheet resistance at $T=4.2$ K caused by the hydrostatic pressure as discussed earlier. The reduction of the classical scattering behaviour, enables the observation of other behaviours of MR. In this case, MR displays a steep increase already for small $H$ and suggests a logarithmic increase at higher $H$. 
5.3 Logarithmic correction to the conductance below 10 K.

To discuss the small increases of $R_s(T)$ below $T=10$ K in excess of $R_{imp}$ we plot the negative conductance difference $-\Delta \sigma(T) = -\left[1/R_s(T)-1/R(T^*)\right]$ versus $\ln(T/K)$ for $T \leq T^*$. The result is shown in Fig. 5.15.

$-\Delta \sigma(T)$ decreases linearly with increasing $\ln(T/K)$. The slope is of the order of $e^2/\pi h$, where $h$ is the Planck constant, which indicates contributions from weak localization (WL) and/or electron-electron interaction (EEI) [62]. WL effects originate from the fact that in a disordered electronic systems, electrons move in a diffusive rather than ballistic nature. That is, electrons do not move in a straight line, but rather experience multiple scattering events by impurities in the sample, which results in a random path. $R_s$ of the system is related to
5.3. Logarithmic correction to the conductance below 10 K.

the probability of the electrons to propagate between two given scattering points in the STO. In classical physics, this probability is simply the sum of the probabilities of a paths connecting the two points. However, in quantum mechanics, the total probability is the summation of the quantum-mechanical amplitudes of all paths rather than the probabilities themselves. Therefore, the correct formula for the probability for an electron to move from a point A to a point B includes the classical part (individual probabilities of diffusive paths) plus the number of interference terms. The interference terms are also present for the particular case A = B. Thus, these interference terms effectively increase the return probability where an electron will "wander around in a circle" than in the classical case, which leads to an increase in the net resistivity. In case of WL, the application of magnetic field leads to a decrease in $R_s$ [59]. However, this was not observed in our MR measurements(see Figs. 5.13 and 5.14). For two dimensional systems (2DES), both WL and EEI lead to a logarithmic dependence of $\Delta \sigma$ on $T$ [63]:

$$\Delta \sigma(T)_{WL} = \alpha r e^2/2\pi^2 \hbar \ln\left(\frac{T}{T_0}\right)$$ (5.7)

and for the electron-electron interaction:

$$\Delta \sigma(T)_{EEI} = \lambda e^2/2\pi^2 \hbar \ln\left(\frac{T}{T_0}\right)$$ (5.8)

where $\alpha r$ and $\lambda$ are dimensionless constants that depend on the material and display values close to unity.

In systems where Rashba spin-orbit interaction is present, which is the case for LAO/STO heterostructures, weak anti-localization (WAL) is expected more likely than weak localization(WL) [37], which leads to a positive correction to the conductance with decreasing $T$, resulting in:

$$\Delta \sigma(T)_{WAL} = -\alpha r e^2/2\pi^2 \hbar \ln\left(\frac{T}{T_0}\right)$$ (5.9)

Hence we expect a drop in $R_s$ with decreasing $T$. However, this was not observed in $R_s(T)$ curves for any pressure, see Fig. 5.15.

In Fig. 5.16, the negative magnetoconductance $-\Delta \sigma(H) = 1/R_s(H)-1/R_s(0)$ is shown for $p=1.64$ GPa at $T=4.2$ K versus magnetic field $\mu_0 H$. 
5.3. Logarithmic correction to the conductance below 10 K.

Figure 5.16: The negative magnetoconductance $-\Delta \sigma(H) = 1/R_s(H) - 1/R_s(0)$ at $p=1.64$ GPa and $T=4.2$ K as a function of magnetic field $\mu_0 H$. The negative magnetoconductance increases with increasing fields. $-\Delta \sigma$ displays a logarithmic field dependence for $\mu_0 H > 1$ T.

For EEI, a magnetic field dependence $-\Delta \sigma^{EEI}(H) = \lambda/2 (e^2/\pi h) G(H)$ is expected. Where:

$$G(H) = \ln(h^*/1.3) \quad \text{for} \quad h^* > 1$$
$$G(H) = 0.084h^{*2} \quad \text{for} \quad h^* < 1$$

with $h^* = g\mu_B\mu_0 H/k_B T$ \[63\] $g$, $\mu_B$ and $k_B$ are the electron $g$ factor, Bohr magneton and the Boltzmann constant, respectively.

At 4.2 K a magnetic field of 5 T corresponds to for $h^* \approx 1$. According to Eq. 5.10, we therefore expect a change in the behaviour from power law to logarithmic for $\mu_0 H > 5$ T. This change in behaviour, however, is observed at $\mu_0 H > 2$ T instead of 5 T. One needs to keep in mind that in Ref. \[64\] spin orbit and spin scattering are not considered, which results in a
5.3. Logarithmic correction to the conductance below 10 K.

Considerable increase in $h^*$, and to $-\Delta \sigma^{EEI}(H) \propto \ln(H)$ at smaller fields [61].

The observed change between 0 and 7 T of the conductance at 1.64 GPa is again of the order of $e^2/\pi h$. Similar changes in $\Delta \sigma(H)$ and $\Delta \sigma(T)$ are obviously caused by comparable changes in $T$ and $H$, i.e., $g \mu_B \mu_0 \Delta H \approx k_B \Delta T$. Hence, the corrections to the conductance below $T^*$ with respect to their $T$ and $H$ dependencies are consistently described by EEI. However, the positive MR($H$) of LAO/STO is often alternatively discussed in the context of WAL [64, 37].

For weak localization, the magnetoconductance of a 2D layer is given by [65]:

$$\Delta \sigma(H_{si}, H_{tr}, H) = \frac{e^2}{h} \left( \frac{3}{2} f(H/H_{tr}) - \frac{1}{2} f(H/H_{si}) \right)$$

(5.11)

where

$$f(b) = \frac{1}{\pi} \left( \psi\left(\frac{1}{2} + \frac{1}{b}\right) - \ln(b) \right)$$

(5.12)

The triplet and singlet states contributions are described by:

$$H_{tr} = 4H_{SO} + H_{\phi}$$

(5.13)

$$H_{si} = H_{\phi}$$

(5.14)

where $\psi$ is the di-gamma function, $H_{\phi}$ is the inelastic field, describing the phase relaxation process by inelastic scattering and the $H_{SO}$ is the spin orbit-field, describing spin relaxation by spin-orbit interaction. If spin-orbit interaction in LAO/STO was absent, this would lead to a positive $\Delta \sigma(H_{\phi}, H) = (e^2/2h)f(H/H_{\phi})$, whereas in case of strong spin-orbit coupling, $\Delta \sigma(H_{\phi}, H) = -(e^2/2h)f(H/H_{\phi})$ becomes negative. The phase coherence length is given by $L_{\phi} = [h/(8\pi e H_{\phi})]^{1/2}$. 
In order to check whether our data are compatible with WAL, we plot (Fig. 5.17) the negative magnetoconductance $-\Delta \sigma(H) = 1/R_s(H)-1/R_s(0)$ versus $\mu_o H$ at $p=1.64$ GPa and $T=4.2$ K. The fit of $-\Delta \sigma(H)$ according to Eq. 5.11 is shown by the solid line. The data can be well described in terms of 2D WAL [62, 65, 64] if the conductance is multiplied by a factor 2.2. The inelastic field $H_\Phi = 75$ mT and a spin-orbit field $H_{SO} > 8T$, deduced from the fit compares well to data reported in Ref. [37]. However, in view of our observations described above, EEI seems to be more likely responsible for the observed magnetoconductance at low $T$. 

Figure 5.17: The negative magnetoconductance $-\Delta \sigma$ versus $\mu_o H$ at $p=1.64$ GPa and $T=4.2$ K. The solid line indicates a fit to Eq. 5.11
Chapter 6

Summary

We have successfully prepared high quality LaAlO$_3$/SrTiO$_3$ heterostructures using pulsed laser deposition technique under optimized parameters. The structural and electrical properties of these heterostructures were investigated by means of atomic force microscope, X-ray, high resolution (aberration corrected) transition electron microscope (HR(AC)TEM). New hardware, including a pressure cell, were designed and built to allow for measurement of the electrical resistivity and magnetoresistance under hydrostatic pressure. The sheet resistance was measured as a function of temperature between 4.2 K and 300 K at ambient and at different hydrostatic pressure up to $p=1.46$ GPa. Magnetoresistance measurements were performed up to 7T on the same samples at different temperature and pressure.

Our study documents the very vital role of donor states and the relative permittivity($\varepsilon_r$) of STO (especially at low $T$) for the transport properties of the 2DES in LAO/STO heterostructures under hydrostatic pressure. For $T > 100$ K, the transport properties are determined by electron-phonon scattering resulting in $R_s \propto T^2$. $R_s$ depends only very little on $p$, due to the small dependence of $\varepsilon_r$ on hydrostatic pressure at this temperature range. However, for $10$ K $< T < 100$ K, $\varepsilon_r$ is highly dependant on both $p$ and $T$, strongly affecting $R_s(T)$ by impurity scattering due to, most likely, to oxygen vacancies. Thermal activation of trapped charge carriers and the para-electric behavior of STO lead to a distinct $T$ dependence of the impurity scattering in this temperature range with a minimum in $R_s(T)$ occurring at $T_{min}$, separating the metallic ($T > T_{min}$) and nonmetallic ($T < T_{min}$) regimes of the 2DES. The strong $p$ dependence of the dielectric permittivity $\varepsilon(p)$ of STO primarily is responsible for the incipient localization of charge carriers, i.e., the strong decrease of $\mu_H$ and increase of $R_s$ with increasing $p$. Hence, it is the $p$ and $T$ dependencies of $N_i(T)$ and $\varepsilon(p,T)$, rather than the Kondo effect, that entail the $R_s(T)$ minimum under pressure. The small negative logarithmic corrections to the conductance
observed for $4.2 \text{ K} < T < 10 \text{ K}$ are attributed to electron-electron interactions.

Our work not only increased our understanding of these systems, but also it opened the door to new interesting experiments. One of which is the application of gate voltage on the sample while under the maximum hydrostatic pressure. The application of the electric field on the sample improves the conductivity by increasing the number of charge carriers in the system. The drop in $\varepsilon$ due to the application of hydrostatic pressure, leads to reduction in the screening of the applied electric field, which helps achieve higher electric field on the carrier using lower voltages than the ones used at ambient pressure. This experiment might help understand the contribution of the charge carrier concentration and the Hall mobility on the sheet resistance, and see if the increase in the sheet carrier concentration can reverse the drop in the Hall mobility caused by the hydrostatic pressure at low temperature.
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