Effect of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ thin films on electron transport at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface by surface acoustic waves

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ABSTRACT

Surface acoustic waves (SAWs) are capable tools for providing mechanical control over the electronic properties of functional materials. Coupling SAWs with the $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) conducting interface is particularly interesting as this interface exhibits extraordinary features, such as high mobility at low temperature, ferromagnetism, and superconductivity below 200 mK. For SAW generation, piezoelectricity is indispensable, and due to lack of that in the LAO/STO system, a 200 nm thick $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ (PZT) film was grown on top of LAO. SAW excitation and propagation was demonstrated on a PZT/LAO/STO multilayer structure. We further employed SAWs in order to transport free electrons confined to the LAO/STO interface, detected as an acoustoelectric voltage at room temperature. Electrical characterization of the interface was carried out by Van der Pauw measurements. We found that having a PZT layer on top of LAO/STO considerably degraded the interfacial conductivity. The degradation became more pronounced at low temperatures. We attribute these effects to the filling of oxygen vacancies due to interlayer oxygen migration, combined with carrier freeze-out at low temperatures.

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I. INTRODUCTION

Charge transport by surface acoustic waves (SAWs) in new classes of low-dimensional materials has been extensively studied in the last few decades, in order to gain new perspectives and broaden the application areas in electronic devices and new technologies,^{1–5} e.g., involving hybrid quantum systems.^{6,7} SAWs are a form of periodic lattice deformations that propagate over the surface of a solid.⁸ As they interact with a piezoelectric medium, an electric potential is produced, thanks to the inverse piezoelectric effect, and the SAWs are accompanied by this potential.⁹ As a result, free carriers are trapped within a “SAW-train” and transported alongside, thus generating an acoustoelectric current/voltage.¹⁰ So far, this approach has achieved a considerable interest in the field of (quantum) acoustics and has been demonstrated in various low dimensional materials and heterostructures such as graphene, transition metal dichalcogenides (TMDs), and semiconductor 2D electron systems (2DESs), due to the close proximity of charges to the surface.^{5,11–14} The generation

of SAWs in a medium using RF electronic signals relies on piezoelectric transduction, typically using inter-digital transducers (IDTs). Most of the time, if the material under study is not intrinsically piezoelectric, the functional material is either deposited on a strong piezoelectric substrate or a layer of a suitable piezoelectric material is deposited on top.^{5,12,15,16} However, such an approach might require fine tuning of deposition parameters in order to prevent any degradation of the electronic properties of the materials.

Our particular interest concerns the coupling of SAWs with mobile carriers at the $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) interface. This interface possesses extraordinary properties such as high charge carrier mobility at low temperatures,¹⁷ ferromagnetism,¹⁸ and superconductivity below 200 mK.¹⁹ A minimum of 4 unit cells (u.c.) of LAO is deposited on STO, as a primary condition to form a two-dimensional electron system (2DES)²⁰ confined to the STO side of the interface. Modulation of these features, sourced by the interfacial electron–lattice interactions resulting from SAW phonons, could play a critical role in the emergence of new physical

phenomena or in providing a better understanding of known phenomena. However, the non-piezoelectric nature of the LAO/STO system is a major limitation. Some claims of induced piezoelectricity at the interface of LAO/STO due to lattice-mismatch mediated strain can be found in the literature.²¹ According to our efforts, however, any such piezoelectricity, if present, does not allow for SAW generation. In a very recent work, we showed SAW generation and acoustoelectric transport at the LAO/STO 2DES.²² The absence of piezoelectricity in the LAO/STO system was overcome by electrostriction under large dc bias applied along with the rf signal to the IDTs. Acoustoelectric transport was demonstrated both at room temperature and at 150 K. However, we found that, below 105 K, SAW generation and consequently the acoustoelectric signal were suppressed due to the crystal phase transition of STO from cubic to tetragonal. In order to overcome this and be able to carry out experiments at much lower temperatures, where interesting physics takes place in the LAO/STO system, in the present work, we deposited $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ (PZT), a strong piezoelectric perovskite-oxide material, on top of an LAO/STO heterostructure. In order to minimize any previously mentioned possible degradation, which might affect the LAO/STO interfacial properties while depositing PZT, the process parameters were finely tuned. After these optimizations, the most suitable layer combinations were chosen and acoustic charge transport devices were fabricated comprising PZT/LAO/STO multilayer structures. IDTs for SAW generation and detection were fabricated on top of the PZT layer. Before PZT deposition, the LAO layer was patterned into a Hall-bar with eight probing arms, in order to carry out acoustoelectric charge transport measurements as well as Hall-effect measurements. We determined the temperature dependent carrier mobility, carrier density, and sheet resistance after PZT deposition as a function of different LAO thicknesses. Our results show a clear effect of the PZT layer on the interfacial electronic properties, which becomes highly pronounced at lower temperatures. Nonetheless, we demonstrate SAW driven electron transport at room temperature, detected as a dc voltage, which is analogous to an acoustic battery.

Devices were fabricated out of PZT/LAO/STO multilayer structures, as shown in Fig. 1(a). A 10 unit cells (u.c.) thick LAO layer was grown on a TiO_2 -terminated (100) single crystalline STO substrate by pulsed laser deposition (PLD), and a 2DES was formed at the interface. The deposition was done within ~ 180 laser pulses from a 1-in. single-crystalline LAO target at 850 °C substrate temperature, under 1×10^{-4} mbar O_2 pressure. Then, the substrate was left to cool down at a rate of 10 °C/min in an O_2 pressure of about 1.7×10^{-3} mbar. The number of deposited LAO layers was counted by an *in situ* reflection high-energy electron diffraction (RHEED) technique. After deposition, the LAO layer was patterned in the shape of a Hall-bar, by applying a dedicated patterning process.²³ Subsequently, a 200 nm thick PZT film was deposited in a different PLD chamber over the entire sample to be used as a piezoelectric layer that is required for SAW excitation. PZT films were deposited at 600 °C substrate temperature, at 2.5 J/cm² laser energy density, and under 10^{-1} mbar O_2 pressure. After deposition, films were cooled to room temperature in a 1 bar oxygen atmosphere and at a cooldown rate of 10 °C/min. IDTs were used in order to generate and detect SAWs. They were fabricated in a delay-line configuration by standard photolithography and e-beam

FIG. 1. (a) Cross-sectional illustration of a PZT/LAO/STO acoustic charge transport device. Signal and ground electrodes of the IDTs are shown as small black and gray squares on top of the PZT layer, respectively. (b) Optical microscope image (top-view) of the device. IDTs are shown in yellow on both sides of the Hall-bar structure (orange on a greenish background). Note that in this figure, a double finger IDT design is shown, while the measurements in this work were done by using a (otherwise similar) single finger IDT design.

evaporation on top of the PZT film. In Fig. 1(b), an optical microscope image of the device is shown. IDT electrodes were designed in the single-finger (SF) configuration with 2.5 μm finger width and 10 μm periodicity, which also determines the wavelength of our SAWs. The IDTs were made out of a Cr/Al/Cr (5 nm/100 nm/5 nm) stack and separated equally, by 100 μm, from both sides of the Hall-bar structure, which was designed to have a width of 10 μm and a length of 300 μm. The numbered Hall-bar arms/pads are separated equally from each other. They are employed to measure SAW driven electron transport through the microchannel, either as dc current (short-circuited condition) or as dc voltage (open condition).

In Fig. 2(a), the SAW transmission characteristics of a PZT/LAO/STO structure are shown. The time gating function of the vector network analyzer (VNA) was used in order to filter out electromagnetic (EM) signals that are transmitted between IDTs at much higher speed. Note that time gating also suppresses certain interference effects resulting from reflected SAWs. The excited peak at 426 MHz is the first harmonic SAW signal, and the next excited peak at 1.2 GHz is the third harmonic of the related SAW mode. Figures 2(b)–2(d) show non-gated transmission, gated transmission, and reflection signals of the third harmonic in a narrower frequency range, respectively. In Fig. 2(b), SAW transmission is observed with double peaks and a strong dip, which resembles the transmission characteristics of a common type of SAW resonator.²⁴ This is due to the large number of fingers in the IDT, resulting in significant internal back-reflection of SAWs, which shows up as a sharp dip in transmission. This could be overcome by decreasing the number of fingers used in the IDT design. However, as the number of fingers decreases, the signal quality becomes worse in terms of intensity and bandwidth.²⁵ There are particular solutions

FIG. 2. (a) S_{12} characteristics of the device. Peaks show the time gated SAW transmission of the first and third harmonic SAW modes through a PZT/LAO/STO multilayer structure, for a $10\text{ }\mu\text{m}$ SAW wavelength. (b) S_{12} characteristics of the third harmonic signal measured without time gating. (c) Time gated third harmonic SAW transmission signal shown in a narrow frequency range. (d) S_{11} (reflection) characteristics of third harmonic SAWs.

that already exist; however, since for our present purposes minimizing reflections is not important, this will not be discussed any further in this report. In Fig. 2(c), the time gated signal of the same SAW transmission is shown.

After the transmission of SAWs had been confirmed, acoustoelectric transport was measured as a dc voltage by connecting a sensitive voltmeter to different contact pads on the Hall-bar channel. The experiments were carried out at room temperature in the dark and in vacuum. A 12 dBm RF input sine wave was applied to the source IDT during the transport experiments, and the frequency span was specified in a narrow window around the third harmonic SAWs excited at 1.2 GHz. First, IDT1 was used as a SAW source and a voltmeter was connected to pads 2 and 6 as indicated in Fig. 3(a). Because these pads are on opposite sides of the Hall-bar, i.e., perpendicular to the direction of SAW propagation, no potential difference

FIG. 3. (a) Acoustoelectric voltage across two contact pads separated perpendicular to the SAW propagation direction. For this particular measurement, contact pads 2 and 6 were used [Fig. 1(b)] with a separation distance of $10\text{ }\mu\text{m}$, i.e., the width of the Hall-bar. (b) Acoustoelectric voltage across contact pads 2 and 3, with a separation of $100\text{ }\mu\text{m}$. IDT1 was used as the SAW input. (c) Same as (b), but with IDT2 used as the SAW input.

was built-up in the specified direction, as expected. In the following measurement, the voltmeter was connected to two pads separated along the direction of SAW propagation, and SAWs were sourced from IDT1. As electrons are pushed toward IDT2, a finite potential difference developed, and this was measured by probing the Hall-bar channel in between pads 2 and 3. At resonance frequency, we measured a dc voltage of about 0.5 nV . When the source IDT was switched to IDT2, the signal changed sign but appeared exactly at the same frequency due to SAW propagation and electron transport in the opposite direction [see Fig. 3(c)].

The results obtained in Figs. 2 and 3 confirm the acoustoelectric coupling between LAO/STO interfacial electrons and the SAWs that propagate over the PZT/LAO/STO multilayer structure. The amplitude of the acoustoelectric voltage may be increased at low temperatures, where the carrier mobility is expected to become higher and thermal vibrations are reduced. This follows from a simple relaxation model¹² according to the expression $V_{AE}/R_{2DES} = -\mu(P_{SAW}/v_{SAW})\Gamma$, where v_{SAW} is the SAW velocity, μ is the LAO/STO interface mobility, Γ is the attenuation coefficient, and P_{SAW} is the power of the SAWs. Due to proportionality between V_{AE} and μ , the acoustoelectric voltage may be expected to increase by increasing the 2DES mobility. In our measurements, we found that the bare LAO/STO interface mobility reaches up to $\sim 600\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at $\sim 10\text{ K}$, whereas it is only $\sim 2\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at room temperature as shown in Fig. 4(b) (orange circles). When an additional PZT film was grown on top of the LAO layer, the sheet resistance and mobility values show an abrupt change at about 50 K , [Figs. 4(a) and 4(b)], while the carrier density is suppressed considerably [Fig. 4(c)]. The results obtained in these figures show that the PZT layer is responsible for degradation of the electronic properties of the LAO/STO interface.

FIG. 4. 200 nm thick PZT films were deposited on LAO/STO samples with 10, 20, and 40 u.c. thick LAO layers. The corresponding data points are shown with squares, triangles, and stars, respectively, in (a), (b), and (c). A separate sample with a 10 u.c. LAO layer was also prepared as a reference; the corresponding data points are shown with circles in the same figures for comparison. Films are unpatterned and the surface area is $5 \times 5 \text{ mm}^2$. (a) Sheet resistance, (b) carrier mobility, and (c) carrier density of the LAO/STO interface as a function of temperature. (d) Effect of PZT deposition conditions, without depositing PZT, on carrier density and mobility of the reference sample with a 10 u.c. LAO layer as a function of temperature. Circles and triangles blue (pink) symbols/lines show carrier density (mobility) before and after the sample was exposed.

Oxygen vacancies, which are a main source of conductivity at the interface, might be filled through an interlayer oxygen migration process²⁶ after PZT is grown on top.

In order to investigate and minimize this effect, we decided to use the LAO layer as an oxygen diffusion barrier, and by increasing the LAO film thickness, we aimed to keep the LAO/STO interface further away from the PZT layer. Unpatterned LAO films with 10, 20, and 40 u.c. thicknesses were grown on STO substrates, and 200 nm thick PZT films were deposited on top of all three samples, except for a reference LAO (10 u.c.)/STO sample. The temperature dependence of R_s (sheet resistance), μ (carrier mobility), and n_s (sheet carrier density) of the interface is determined by a Van der Pauw method and presented in Figs. 4(a)–4(c), respectively. In Fig. 4(a), the effect of the PZT layer on the sheet resistance is clearly visible as the samples with PZT are compared with the reference sample. When the interface was separated further from the PZT layer by increasing the thickness of the LAO film, the room temperature resistance of the interface became much larger. At $\sim 50 \text{ K}$, a sharp upturn of R_s was observed for every sample, except for the reference sample. At much lower temperatures, samples with 20 and 40 u.c. thick LAO became completely insulating, inhibiting any electronic transport measurements, while the resistance variation saturated for the sample with 10 u.c. thick LAO. Consistent behavior was also obtained for interface mobility, as shown in Fig. 4(b). At 50 K, the interface mobility showed a broad maximum for the samples with PZT top layers.

The results shown in Figs. 4(a) and 4(b) are clearly not in good agreement with our initial goal. Separating the interface

further from the PZT layer, by increasing the thickness of the LAO film, did not prevent the effects due to oxygen diffusion. On the contrary, an unexpected increase in the resistance occurred.^{27,28} In Fig. 4(c), the strong suppression of the carrier density after PZT thin film deposition is very dominant and is also independent of the separation between the PZT film and the LAO/STO interface. This might indicate that the increase in the LAO layer thickness is not sufficient to prevent oxygen diffusion.

For further investigations, another set of measurements was carried out on a separate sample. A 10 u.c. LAO film was deposited as a reference sample, without PZT deposition on top, and characterized with the same Van der Pauw method [Fig. 4(d), data points represented by circles]. After that, the sample was exposed to the PZT film deposition conditions (high deposition temperature and high oxygen pressure for 1 h), but the PZT deposition was prevented by protecting the sample surface using a mechanical shutter. The same characterization procedure was repeated [see Fig. 4(d), data points represented by triangles]. As is evident from Fig. 4(d), the effect on the interfacial carrier density and mobility measured before and after the process is almost negligible in a wide temperature range. Therefore, we conclude that the deposition process, even though it is carried out under relatively high oxygen pressure (10^{-1} mbar), does not cause noticeable degradation in our devices. On the other hand, the effect is visible only when a PZT film is on top of LAO/STO. Besides interlayer oxygen diffusion mentioned above, this also might be related with spontaneous polarization of PZT. Depending on the polarization direction, charge carriers at the LAO/STO interface may be depleted.^{29,30} In addition, disorder in the spontaneous polarization

due to domain formation could lead to potential fluctuations and, therefore, increased scattering of carriers.

The degradation at the LAO/STO interface after deposition of PZT can be linked with previous work done by Huijben *et al.* where they explained a similar effect in a different material system by interfacial oxygen vacancy filling through an oxygen exchange process between the layers in the multilayer structure.²⁶ This would explain why we did not observe any effect in Fig. 4(d), while a strong degradation was measured for samples with a PZT layer on top. It is also claimed in the same work that such an exchange process could give rise to an increased mobility, as the oxygen vacancies are known to be scattering centers. However, our results show the opposite behavior. As seen in Fig. 4(b), below 100 K, the interfacial mobility values show a dramatic decrease. This behavior is well explained by a carrier freeze-out phenomenon, proposed by Liu *et al.* Since the number of carriers is highly reduced after oxygen vacancy filling, the band due to oxygen vacancies narrows and separates from the conduction band, such that the remaining carriers are trapped at a certain temperature and hence the interface suddenly becomes highly insulating.³¹ An additional explanation can be given according to two separate studies by Bell *et al.* and Herranz *et al.* Both studies showed an upturn in the sheet resistance and a considerable decrease in mobility at around the same temperature, as the number of LAO layers exceeded ~ 10 u.c.^{27,28} Since the LAO thickness in our samples was increased up to 40 u.c., which is outside the usual range for obtaining a uniform 2DEG at the LAO/STO interface, our results could also be (partly) explained in a similar way.

II. CONCLUSION

Although our results clearly demonstrate acoustoelectric coupling between SAWs and charge carriers at the LAO/STO interface, it is found that deposition of PZT on top of LAO/STO is unfavorable. We measured a surprising transition of interfacial conductivity into the insulating regime below 100 K. This could be explained by charge carrier freeze-out after the interfacial oxygen vacancies are filled during (or after) the deposition of PZT due to interlayer oxygen diffusion. To allow for acoustoelectric transport at low temperature, a possible solution could be the deposition of a PZT thin film only underneath the IDTs. By this way the 2DES region may be protected, while the PZT will still provide the piezoelectric platform required to excite SAWs. However, fabrication of such a device may be quite challenging since it involves the development of an appropriate patterning technique that does not degrade the LAO/STO interface, which is not within the scope of this work. However, we propose that the development of a damage-free fabrication process for such a device can ultimately avoid the aforementioned problems.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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