Superconductivity at the LaAlO₃/SrTiO₃ interface

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Abstract. We report on the structural characterization of LaAlO₃/SrTiO₃ interfaces and on their transport properties. LaAlO₃ films were prepared using pulsed laser deposition onto ${\rm TiO_2}$ terminated (001) ${\rm SrTiO_3}$ substrates inducing a metallic conduction at the interface. Resistance and Hall effect measurements reveal a sheet carrier density between 0.4 and $1.2 \cdot 10^{14} \, \mathrm{electrons/cm^2}$ at room temperature and mobility of $\sim 300 \, \mathrm{cm^2 V^{-1} s^{-1}}$ at low temperatures. A transition to a superconducting state is observed at a temperature of ~ 200 mK. The superconducting characteristics display signatures of 2D superconductivity.

1. Introduction

Ohtomo and Hwang discovered in 2004 that metallic conduction occurs at the interface between two insulating materials, LaAlO₃ (LAO) and SrTiO₃ (STO) [1]. This discovery has generated a lot of interest and motivated extensive theoretical and experimental work aimed at investigating the physics of complex oxide interfaces. In such systems, novel electronic properties have been predicted [2] and recently observed [3,4]. In the specific case of the LAO/STO interface, magnetic effects [5], superconductivity [6] and a rich electronic phase diagram [7] have been reported. The mechanism leading to the formation of a mobile electron gas at the LAO/STO interface remains today a highly debated issue. A possible explanation is the so-called 'polar catastrophe' scenario. According to this interpretation, the origin of this phenomenon is linked to a polar discontinuity occurring at the LAO/STO interface between the polar (001) LAO atomic planes and non-polar (001) STO planes [8]. When the polar LAO atomic planes are stacked on top of each other, an electric potential develops as the film thickness increases. Above a critical thickness, an electronic reconstruction may take place leading to a charge transfer at the interface, reducing the electrostatic energy and creating a two dimensional electron gas. Experimental evidences supporting this scenario have been reported in ref. [9], where the electron gas and metallic conduction have only been observed for LAO films at least 4 unit cells (u.c.) thick. The confinement of the electron gas has been estimated from analyses of the superconducting properties [10] and by room temperature conductive atomic force measurements [11]. These experiments lead to an estimate of the thickness of the electron gas of a few nanometers. In this short paper we report on the structural characterization of LAO/STO structures and describe their transport properties in the normal and superconducting states. We analyze the superconducting behavior in the framework of the Berezinskii-Kosterlitz-Thouless (BKT) theory of 2D superconductivity.

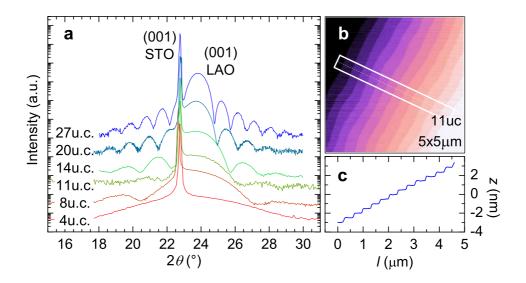


Figure 1. Structural characterization of LAO thin films. (a) X-ray $\theta - 2\theta$ diffractograms around the (001) LAO and STO reflections for LAO thin films with different thicknesses. (b) AFM topography performed on an 11 u.c. LAO thick film and (c) a height profile averaged over the width of the area delimited by the white lines. The step-and-terrace structure reflecting the miscut of the STO substrate is clearly visible.

2. Experimental details

Conducting interfaces were prepared depositing more than 3 u.c. of LAO on top of (001) SrTiO₃ single crystals with TiO₂-terminated surfaces. The films were grown by pulsed laser deposition (PLD) at 800 °C in 1·10⁻⁴ mbar of O₂ with a repetition rate of 1 Hz. The fluence of each laser pulse was 0.6 J/cm². After the LAO deposition, each sample was annealed in 200 mbar of O₂ at about 600 °C for one hour and cooled down to room temperature in the same O₂ background pressure. The growth was monitored in situ using reflection high energy electron diffraction (RHEED) which allows a control of the thickness with sub u.c. precision [12]. Samples with different thicknesses of LAO were systematically characterized, analyzing the surface morphology by atomic force microscopy (AFM) and crystalline quality by X-ray diffraction (XRD). The samples were then patterned in a geometry suitable for 4 point transport measurements. The patterning technique is based on the fact that only regions covered by more than 3 u.c. are conducting [9]. The LAO thickness was thus reduced using Argon ion milling down to 2 u.c. in specific regions of the samples while protecting the transport channels with photoresist. Once the conducting channels were defined, the transport properties were investigated in a dilution cryostat.

3. Results

Figure 1a displays $\theta-2\theta$ X-ray diffractograms close to the (001) LAO and STO reflections for samples with different LAO thicknesses. As can be seen in the Figure, the period of finite size Fresnel oscillations around the diffraction peak changes with the LAO film thickness and allows the thickness of the LAO layer to be determined. The thickness obtained from the XRD data analyses is found to agree within one unit cell with the thickness deduced from the RHEED intensity oscillations. Figure 1b shows a typical AFM topography measured on an 11 u.c. LAO sample. We observe that the steps-and-terraces structure of the substrate is reproduced on the surface of the film. The step height is about 4 Å, corresponding to one u.c of LAO, as shown in Figure 1c.

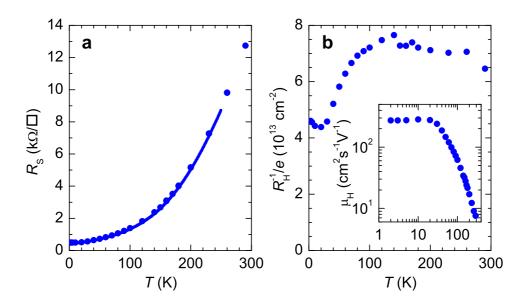


Figure 2. Transport properties in the normal state for a LAO/STO structure. (a) Sheet resistance as a function of temperature for a 4 u.c. thick LAO film grown onto a (001) STO substrate. The continuous line represents the resistance measurement during the cooling while the dots are measurements performed stabilizing the temperature during the heating cycle. (b) Inverse sheet Hall constant and mobility (inset) versus temperature for the same sample.

Reciprocal space maps (RSM) were performed to inspect the structural coherence of the LAO films grown atop the STO substrates. Measurements of RSM around the (103) diffraction peaks for film and substrate reveal that the in-plane lattice parameter of the LAO layer is strained to the one of STO. This demonstrates that LAO thin films grow pseudomorphically on STO substrates.

Transport measurements reveal that these heterostructures have a typical sheet resistance R_S between 10 and $30\,\mathrm{k}\Omega/\mathrm{square}$ at room temperature. Most of the samples display a small photoelectric effect ($\Delta R/R < 5\%$) under ambient light illumination. As shown on Figure 2a, resistance measurements indicate a metallic behavior down to 4 K, with a residual resistivity ratio of the order of 25. Figure 2b displays the inverse sheet Hall constant as a function of temperature for the same sample. Considering a single band model, we estimate a sheet carrier density of the order of $7\cdot10^{13}$ electrons/cm² above 100 K. From sample to sample, the sheet carrier density varies between 0.4 and $1.2\cdot10^{14}$ electrons/cm², without correlation with the thickness of the LAO layer. As can be noted in the Figure, below 100 K, the inverse sheet Hall constant decreases as the temperature is reduced. Such a behavior has previously been observed in Nbdoped SrTiO₃ thin films [13] and attributed to multiband conduction. Although this issue is still controversial, it is known that doped SrTiO₃ possesses three degenerate bands for Ti 3d electrons [14]. This degeneracy is lifted by the structural phase transition occurring at $\sim 105\,\mathrm{K}$. The inset of Figure 2b shows the mobility versus temperature which reaches a value of $\sim 300\,\mathrm{cm}^2/\mathrm{Vs}$ at low temperatures.

Measurements of the resistance at low temperatures reveal that this electron gas condenses into a superconducting state. Figure 3a shows the sheet resistance versus temperature for a 4 u.c. thick LAO film. As can be seen, a superconducting transition occurs at a temperature of $\sim 220 \text{ mK}$. From magnetotransport measurements with the magnetic field applied perpendicular to the interface, we have estimated $H_{c2}(0 \text{ K})$ to be $\sim 90 \text{ mT}$ [6] which leads to a superconducting coherence length $\xi(0)$ of about 60 nm. As the thickness of the electron gas is below 10 nm [6, 11],

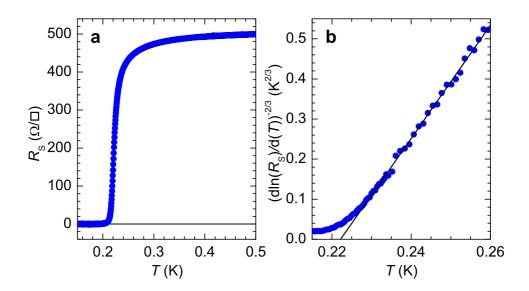


Figure 3. Transport properties at low temperatures.(a) Sheet resistance versus temperature for a 4 u.c. sample. (b) $(d \ln R_S/dT)^{-2/3}$ versus temperature revealing the linear behavior expected for a BKT transition: the extrapolation of the linear part to zero resistance leads to an estimate for T_{BKT} of about 222 mK.

we expect the system to behave as a 2D superconductor possibly with footprints of a Berezinskii-Kosterlitz-Thouless (BKT) transition [15, 16].

In the BKT scenario, the resistance as a function of temperature is expected to follow the relation: $R = R_{\circ} \cdot \exp[-b(T/T_{BKT}-1)^{-1/2}]$, where T_{BKT} is the BKT transition temperature and b a material dependent parameter. To estimate T_{BKT} and b we rewrite the expression as $(d \ln R/dT)^{-2/3} = (2/b)^{2/3}(T - T_{BKT})$. This form has the advantage that b and T_{BKT} can be estimated from the characteristic linear behavior above T_{BKT} , without having to determine R_{\circ} . This analysis is shown in Figure 3b, where we notice that the resistance measurements are consistent with the BKT behavior. The rounding of the transition at the lowest temperatures is attributed to a size effect. The linear extrapolation to R=0 leads to T_{BKT} =222 mK.

4. Conclusion

We have discussed the growth and characterization of LAO films grown onto (001) STO TiO₂-terminated substrates. With the preparation conditions described above, we observe that only structures with a LAO thickness greater than 3 u.c. are conducting. We have shown that the electron gas present at these LAO/STO interfaces condenses into a superconducting state displaying properties expected for a 2D superconductor.

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