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ORIGINAL PAPER

Optical Study of Tetragonal Domains in $LaAlO_3/SrTiO_3$

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Abstract Scanning superconducting quantum interference device (SQUID) measurements recently revealed enhanced channels of conductivity at the conducting $LaAlO_3/SrTiO_3$ (LAO/STO) interface (Kalisky et al. Nat. Mater. **12**, 1091 2013). The orientation of the channels and their thermal behavior suggest that they originate as a consequence of the STO tetragonal domain formation which sets in below ∼105 K. In this work, we use polarized light microscopy to acquire images of the tetragonal domains in the same group of LAO/STO samples. We looked at the configuration of the domains (orientation and spacing) and followed their behavior as a function of temperature and back-gate voltage. The optical data agrees with the electrical behavior mapped magnetically. This direct and independent study of the domain structure confirms that the channel-like conductivity in LAO/STO is due to the STO tetragonal domain structure and emphasizes the importance of STO physics to the interfacial properties. These results demonstrate how small structural changes in perovskite crystals strongly influence the electronic characteristics of heterostructures.

Keywords LaAlO3/SrTiO3 · Polarized light microscopy · Tetragonal domains

1 Introduction

Although $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) are both insulating and nonmagnetic complex oxides, the interface between them exhibits a list of interesting physical phenomena, such as quasi-two-dimensional electron transport with high electron mobility [2], two-dimensional superconductivity at low temperatures [3–7], and electric field-tuned metal insulator and superconductor insulator phase transitions [4, 5, 8, 9]. Bulk magnetization and magnetoresistance measurements also suggest some form of magnetism, depending on the preparation conditions [7, 10–12] and suggest a tendency towards nanoscale electronic phase separation [11]. The nature of conductivity, superconductivity, and magnetism observed in LAO/STO has been the subject of intense research in the past several years.

Local studies of the interface revealed crucial information about the interface physics. Direct magnetic imaging with scanning superconducting quantum interference device (SQUID) microscopy found that submicron ferromagnetic patches coexist with the superconductivity [13] and a critical thickness for the magnetism [14], similar to

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the conductivity which appears only after growing 4-unit cells of LAO [8]. By local mapping of the stray magnetic fields associated with current flow at the interface with scanning SQUID, Kalisky et al [1] revealed that the current modulates over channels with enhanced current flow. Using a scanning single-electron transistor, Honig et al. [15] detected a striped phase along the crystallographic axes and showed that the distribution of stripes depends on back-gate voltage.

Figure 1, adapted from Kalisky et al [1], illustrates the channel-like conduction. These data and especially the behavior of current channels with thermal cycling (Fig. 1f–g) suggest that there is a strong connection between the well-known STO structural transition [16] and the way the current flows at the interface.

STO is a centrosymmetric perovskite material at room temperature that exhibits structural phase transition at 105 K, from cubic to tetragonal [16]. In the tetragonal phase, the crystal unit cells elongate and become rectangular prisms, in one of the three possible directions. This leads to the formation of large domains with a configuration that minimizes the landscape of strain in bulk STO. Such domain patterns have been observed in bulk STO using polarized optical microscopy 50 years ago [17, 18].

In this work, we present an optical study of the tetragonal domains in several LAO/STO samples. The independent view of the domain configuration and behavior with thermal cycling and back-gate voltage provides support to the suggestion that the channel conduction originates in the STO tetragonal domain structure.

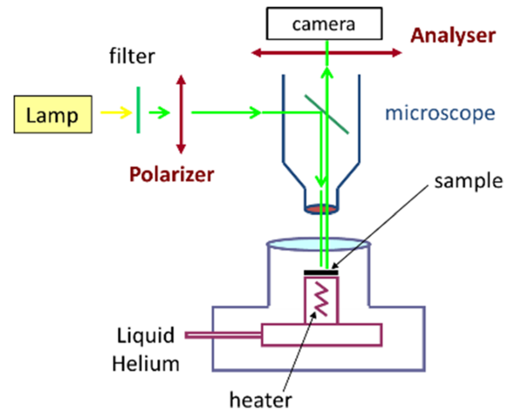

Fig. 2 Illustration of polarized light microscope setup

2 Experimental

Four samples were measured in this work: one unpatterned sample of 5 unit cells (uc) of LAO on TiO_2terminated STO (001) unpatterned and three patterned samples using an AlO_x hard mask (one 5 uc and two 10 uc of LAO). The samples were prepared in a similar manner to Bell et al. [5]. For polarized optical microscopy, we used a constant helium flow optical cryostat, as illustrated in Fig. 2. The samples were held at temperature of 10 K and observed through antireflection-coated fused silica window. Light from a 100-W mercury lamp was linearly polarized and then reflected off the surface of the sample, through the second polarizer

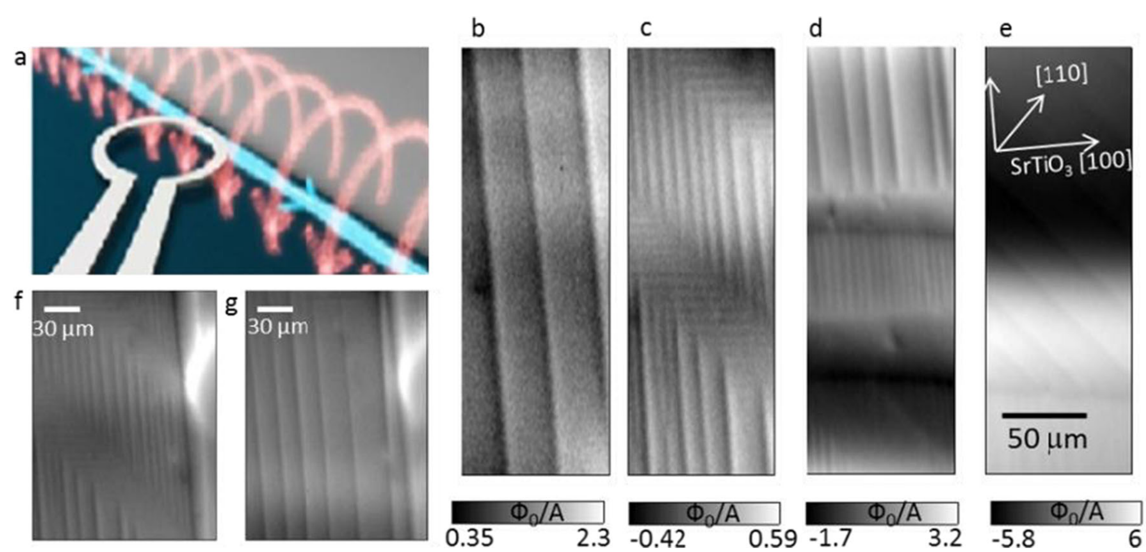

Fig. 1 Scanning SQUID images of LAO/STO channel-like conduction, adapted from Kalisky et al [1]. **a** Illustration of the scanning SQUID mapping of the current flow. The SQUID's pickup loop captures the magnetic flux that is generated by the current flowing in the interface. **b–e** Magnetic flux images from current flowing through four 250 μm × 85 μm regions of a 5 mm × 5 mm sample. The *dark* and *light lines next to each other* correspond to the dipolar feature expected for the perpendicular to interface component of the magnetic field from a wire-like current. **f–g** The configuration of channels changes after cycling the temperature through the 105-K structural transition. **f** Flux map at 4.2 K after cooling down from room temperature. **g** The same area scanned at 4.2 K after cycling to 125 K

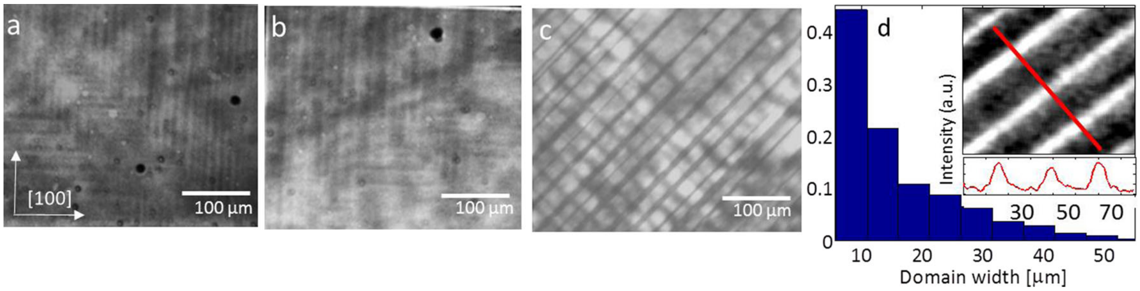

Fig. 3 Polarized microscopy images of domain structure in a 5-unit-cell LAO/STO sample, measured at 10 K. **a**–**b** Domains in [100] and [010], imaged with parallel polarizers. **c** [110] and [−110] domains, imaged with crossed polarizers. **d** The fraction of 777 domains at each width. The inset shows a line cut through the domains

(analyzer) and to the camera. Domain walls along the crystallographic ([100] and [010]) axes were imaged without polarizers, and the [110] and [−110] domains were imaged with nearly perpendicular polarizers.

3 Results and Discussion

We imaged the domain configurations in several samples at different temperatures and back-gate voltages. Figure 3 shows typical data taken in a 5-uc sample with domains in all crystallographic orientations with various widths. The domains observed are in the STO bulk, as can be seen in Fig. 3c where [110] domains are in focus and [−110] domains are out of focus. Figure 3d describes the distribution of domain widths. Few domains are as large as 50 μm, but almost 50 % are smaller than 10 μm wide. This distribution matches the distribution of current channels observed by scanning SQUID microscopy [1].

To further investigate the relationship between current channels and STO domain configuration, we examined the temperature and back-gate dependence of the domains. Figure 4a–c shows optical images of the sample during thermocycling. The domains disappear above 105 K, as

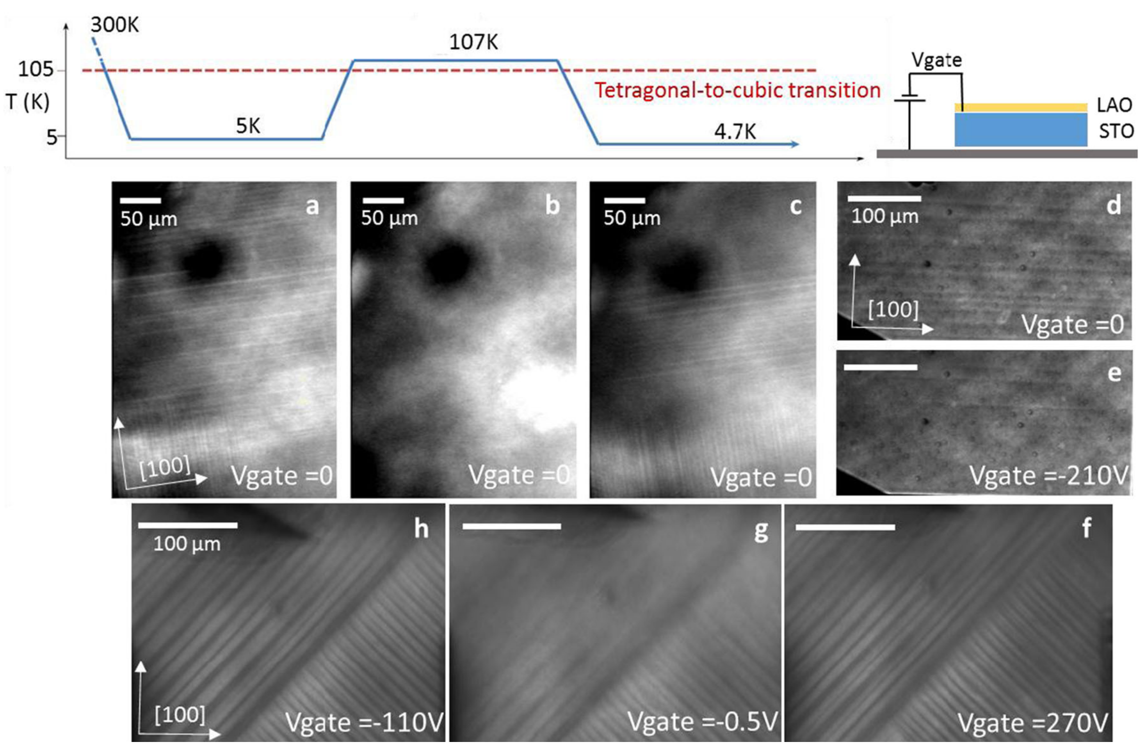

Fig. 4 Optical images of LAO/STO domain structure. **a**–**c** At different cooling cycles. **a** Configuration at 5 K before thermal cycle. **b** No domains observed above the phase transition temperature. **c** Different domain configurations observed in the next cooldown. **d**–**e** Gate voltage dependence of [100] and [010] domains. **d** Without gate voltage applied. **e** Domain structure in the presence of −210 V back-gate voltage. **f**–**h** Gate dependence of [110] and [−110] domains. **d**–**h** Taken at 10 K

expected, and return in a different configuration after a new cooldown through the structural transition. This expected behavior of the domains with temperature resembles the behavior of current channels with temperature, as shown in Fig. 3 in the study of Kalisky et al. [1].

The domain configuration changes with the back-gate voltage in the following way. After the initial cooldown, the [100] and [010] domains are distributed everywhere, but as the back-gate voltage is reduced, they move towards the edges of the sample. Figure 4d shows a 400 μm × 200 μm region covered with [100] domains with no gate applied, and Fig. 4e shows that over half of the domains cleared with gate voltage of −210 V. Figure 4g, h demonstrates that the [110] and [−110] domains behave differently. They are less dominant around 0 V and have maximal coverage at higher or lower back-gate voltages. The behavior that we observed optically agrees with the gate dependence of the [100] striped phase observed by scanning single-electron transistor [15] and the [110] domains observed by scanning SQUID in Kalisky et al. [1].

The goal of this work was to confirm the explanation of the current channels and striped domains observed in the study of Kalisky et al. [1] and Honig et al. [15], by direct imaging. Our results show that the appearance of tetragonal domains and their behavior with temperature and back gate matches the behavior of LAO/STO current channels observed by scanning SQUID microscopy and the gate dependence observed by scanning single-electron transistor. This supports the suggestion of crystallographic domain structure of STO as the origin of the current channels. The microscopic origin of enhanced current flow on domain walls, as discussed in the study of Kalisky et al. [1] and Honig et al. [15], still requires further investigation.

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