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Concomitant appearance of conductivity and superconductivity in (111) LaAlO₃/SrTiO₃ interface with metal capping

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In epitaxial polar-oxide interfaces, conductivity sets in beyond a finite number of monolayers (ML). This threshold for conductivity is explained by accumulating sufficient electric potential to initiate charge transfer to the interface. Here we study experimentally and theoretically the LaAlO₃/SrTiO₃(111) interface where a critical thickness, t_c , of nine epitaxial LaAlO₃ ML is required to turn the interface from insulating to conducting and even superconducting. We show that t_c decreases to 3ML when depositing a cobalt over-layer (capping) and 6ML for platinum capping. The latter result contrasts with the (001) interface, where platinum capping increases t_c beyond the bare interface. Our density functional theory calculations confirm the observed threshold for conductivity for the bare and the metal-capped interfaces. Interestingly, conductivity appears concomitantly with superconductivity for Metal/LaAlO₃/SrTiO₃(111) interfaces in contrast with the Metal/LaAlO₃/SrTiO₃(001) interfaces where conductivity appears without superconductivity. We attribute this dissimilarity to the different orbital polarization of e'_g for the (111) versus d_{xy} for the (001) interface.

I. INTRODUCTION.

The interface between LaAlO₃ and SrTiO₃ exhibits two-dimensional conductivity [1], superconductivity [2], magnetism [3–8], metal-insulator transition [9], tunable Rashba spin-orbit interaction [10, 11], quantum Hall states [12, 13], and one-dimensional conductivity [14, 15]. While the LaAlO₃/SrTiO₃(001) interface has received significant scientific attention, the (111) interface remains less explored.

The Ti atoms in SrTiO₃ form triangular layers along the [111] direction [16, 17]. Due to the trigonal symmetry the degeneracy of the t_{2g} manifold is lifted, splitting into a_{1g} and e'_g orbitals that might lead to topologically non-trivial states in SrTiO₃ quantum wells [18] or to exotic superconductivity [19].

While different scenarios for the formation of conductivity have been proposed at the (001) interface e.g. for amorphous films [20–22], in epitaxial films four monolayers of LaAlO₃ are needed for the formation of a two-dimensional electron system (2DES) at the LaAlO₃/SrTiO₃(001) interface [9]. For the (111) interface, the critical thickness for conductivity is nine monolayers (ML) [16].

Transport properties of the LaAlO₃/SrTiO₃ (111) interface indicate a six-fold symmetry [23]. The (111) 2DES also exhibits superconductivity [24], with a link between superconductivity and spin-orbit interaction [25]. Notably, upon carrier depletion with negative gate voltage, superconductivity transitions into a Bose-insulating state [26]. This behavior contrasts with the (001) interface where a weaker insulating state is observed for negative gate biases [27, 28].

For spin injection and low voltage transistor applications, the barrier produced by the minimal four monolayers of LaAlO₃ required for conductivity at the bare (001)

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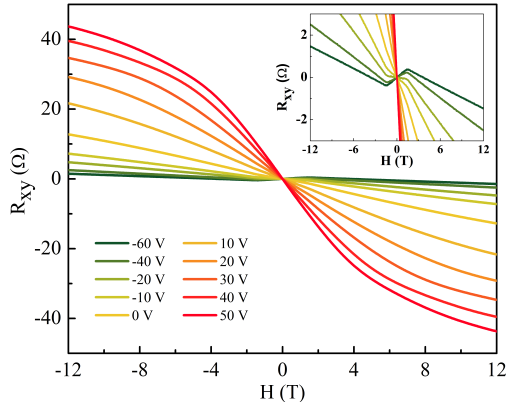


FIG. 1. Transverse resistance of $\text{AlO}_x/\text{Co}/\text{LaAlO}_3/\text{SrTiO}_3(111)$ for 4 ML LaAlO_3 as a function of a perpendicular magnetic field at different gate voltages at 2K. The inset focuses on the negative gate voltage regime. The observed anomalous-Hall signal demonstrates the predominance of the Co layer properties in this regime. This is in contrast to the higher carrier density regime where the 2DES dominates.

interface or by the nine monolayers at the (111) interface is relatively strong. First principles calculations [29], and experimental studies of various metal capping on (001) interfaces (Metal/ $\text{LaAlO}_3/\text{SrTiO}_3$) [30, 31] show that the critical thickness for the onset of conductivity, t_c , can be reduced relative to the bare interface and that t_c increases with the metal work function.

Here we study the problem of the critical thickness for conductivity for (111) interfaces both experimentally and theoretically. We have also expanded our experimental research to the superconducting properties of both (001) and (111) Metal/ $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces. We find that upon capping the $\text{LaAlO}_3/\text{SrTiO}_3(111)$ interface with cobalt (Co) and platinum (Pt), t_c is reduced from $t_c(\text{bare})=9$ ML to $t_c(\text{Co})=3$ ML and $t_c(\text{Pt})=6$ ML. Furthermore, once the (111) interface becomes conducting, it also becomes superconducting at low temperatures. This contrasts with the (001) interface where $t_c(\text{Pt}) > t_c(\text{bare})$.

Concomitant density functional theory calculations with a Hubbard U term (DFT+ U) confirm the reduction of the critical thickness upon metal capping and indicate that conductivity at the (111) interface arises due to bands with e'_g orbital polarization. We conjecture that these bands are also responsible for superconductivity. This is in contrast with the (001) interface where the d_{xy} and the d_{yz}, d_{xz} bands are split due to their different effective masses along the direction of the confining potential (z -direction) [32]. Spin-orbit coupling mixes them together [33] resulting in a lower-energy, non-superconducting band, and a higher-energy, mobile band, which is responsible for superconductivity [17, 34].

II. METHODS

A. Experimental methods

Epitaxial LaAlO_3 films with different thicknesses were grown on Ti, and TiO_2 terminated, atomically smooth (111), and (001) SrTiO_3 substrates respectively at an oxygen pressure of 1×10^{-4} Torr and temperature 780°C using pulsed laser deposition. The detailed growth conditions and characterizations can be found in reference [25, 35]. The thickness was in-situ monitored by reflection high energy electron diffraction (RHEED) (see supplementary information Figure S1). The samples were then transferred to a metal deposition chamber (e-beam for the Co and Ag and Sputtering or e-beam for the Pt) where they were pre-annealed for two minutes at 200°C and at a pressure of 1×10^{-8} Torr to remove surface contaminants [36]. Metallic layers of ≈ 3 nm of platinum (Pt), silver (Ag), or cobalt (Co) were deposited at room temperature. For the Cobalt, we used an additional 3 nm AlO_x to prevent oxidation. Wire bonding was used to connect to the sample electrically.

B. Theoretical modeling

Density functional theory (DFT) calculations were performed on thin LaAlO_3 films on (111)-oriented SrTiO_3 , using the projector augmented wave (PAW) method [37] as implemented in the VASP code [38]. The generalized gradient approximation was used for the exchange-correlation functional, as parametrized by Perdew, Burke, and Ernzerhof [39]. Static correlation effects were considered within the DFT+ U formalism [40], employing $U = 3$ eV for the Ti $3d$ orbitals, in line with previous work [41–43]. The LaAlO_3 thin films on $\text{SrTiO}_3(111)$ were modeled in the slab geometry with two symmetric surfaces to eliminate spurious electric fields. The $\text{LaAlO}_3/\text{SrTiO}_3(111)$ slabs contain 7 monolayers (ML) of SrTiO_3 (substrate) and 3-9 ML of LaAlO_3 on both sides of the substrate. Additionally, in order to assess the role of metallic contacts, a Pt and Co ML was added on top of the LaAlO_3 film. The modeled slabs contain ~ 60 atoms for 3 ML LaAlO_3 and ~ 120 atoms for 9 ML LaAlO_3 , depending on surface termination and metal capping. The lateral lattice constant of the modeled $\text{LaAlO}_3/\text{SrTiO}_3(111)$ slabs was fixed to the SrTiO_3 substrate ($\sqrt{2}a \times \sqrt{2}a$) lateral unit cell with $a = 3.905$ Å. A vacuum region of 15 Å was adopted to minimize the interaction between the slab and its periodic images. A cutoff energy of 600 eV was used to truncate the plane-wave expansion and a Γ -centered k -point mesh of $12 \times 12 \times 1$ to sample the Brillouin zone (BZ). The atomic positions were fully optimized taking into account octahedral rotations and distortions until the forces on all atoms were less than 0.01 eV/Å and the change in total energy was less than 10^{-6} eV. Spin polarization was also considered in the DFT+ U calculations to account for possible mag-

netic moments of the Ti 3*d* electrons and the metal capping layer.

III. RESULTS AND DISCUSSION

A. Experimental results

The transport measurements were performed on the samples with metal capping. In this configuration, the measured resistance is either a parallel combination of the metal cap resistance and the 2DES at the conducting LaAlO₃/SrTiO₃ interface or only the metallic cap in the absence of 2DES. We demonstrate this by measuring the transverse resistance R_{xy} , i.e., the Hall signal of the AlO_x/Co/LaAlO₃/SrTiO₃(111) interface for 4 LaAlO₃ ML, as shown in Fig. 1. While for positive gate voltage, the 2DES dominates and exhibits signal resembling the LaAlO₃/SrTiO₃(111) interface [25] when depleting the 2DES by negative gate voltage, the contribution of Co predominates as manifested in an anomalous-Hall signal, confirming the presence of two parallel channels for conduction. This behavior is similar to (001) with cobalt capping [31].

The transport studies conducted on the AlO_x/Co/LaAlO₃/SrTiO₃(111) interface show a reduction of the LaAlO₃ critical thickness for the onset of 2DES conductivity from 9 LaAlO₃ ML (for the bare interface) to 3 ML (with Co capping). In Fig. 2 (a) we show the sheet resistance of AlO_x/Co/LaAlO₃/SrTiO₃(111) as a function of LaAlO₃ thickness at 40 K. For LaAlO₃ thickness below 3 ML, the resistance increases by nearly a factor of five. This indicates that 3 ML is the critical thickness of LaAlO₃ for the onset of conductivity with Co capping ($t_c(\text{Co})=3$ ML). To verify that a 2DES is formed parallel to the metallic layer, we measured the resistance versus back gate voltage. For a thin metallic layer parallel to a 2DES, one expects gate-dependent resistance due to the dominating contribution of 2DES. On the other hand, in the absence of a 2DES parallel to a metallic layer, we expect the gate dependence of the resistance to be immeasurably small due to the substantial carrier density in the metal. Fig. 2 (b) shows the gate dependence of AlO_x/Co/LaAlO₃/SrTiO₃(111) for 1 and 3 LaAlO₃ ML. For the sample with a single LaAlO₃ ML, the normalized resistance ($R/R_{(-60V)}$) is flat as a function of gate voltage, suggesting the absence of 2DES at the LaAlO₃/SrTiO₃ interface. For 3 ML LaAlO₃, the data show a significant gate dependence, suggesting the formation of a 2DES at the LaAlO₃/SrTiO₃(111) interface. We conclude that t_c becomes 3 ML upon Co capping for (111) interface.

Previous studies on the metal-capped LaAlO₃/SrTiO₃(001) interface show that t_c increases with the work function of the metal-capping layer [29]. To understand the role of the work function in (111) interfaces, we carried out experiments with Pt capping. The work function of platinum is generally

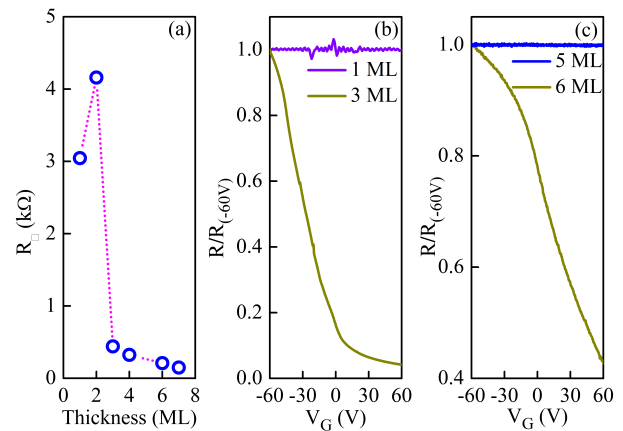


FIG. 2. (a) The sheet resistance (R_{\square}) of AlO_x/Co/LaAlO₃/SrTiO₃(111) at 40 K as a function of LaAlO₃ thickness, an abrupt drop in the resistance at 3 ML indicates that this is the critical thickness for the formation of 2DES at the SrTiO₃/LaAlO₃ interface. Note: The measured samples were not patterned, the geometrical factor for the sheet resistance calculation is within an error bar of $\pm 10\%$. (b) The gate dependence of the normalized resistance ($R/R_{(-60V)}$) for AlO_x/Co/LaAlO₃/SrTiO₃(111) with LaAlO₃ thickness of 1 and 3 monolayers. (c) The gate dependence of normalized resistance ($R/R_{(-60V)}$) for Pt/LaAlO₃/SrTiO₃(111) for LaAlO₃ thickness of 5 and 6 monolayers.

higher than that of Co [44]. Surprisingly, we found that, unlike the (001) interface, Pt capping reduces the critical thickness from $t_c(\text{bare})=9$ ML to $t_c(\text{Pt})=6$ ML. This is demonstrated in Fig. 2(c), where we show the gate dependence of Pt/LaAlO₃/SrTiO₃(111) for 5 and 6 LaAlO₃ ML. The absence of gate dependence for 5 ML and the strong gate dependence for 6 ML suggests that $t_c(\text{Pt})=6$ ML. We interpret the saturation of the resistance at negative gate voltage as a result of depletion of the 2DES and dominance of the metal capping layer (see figure S2 in the supplementary information). In Figure S3 (a) (Supplementary information), we also show the gate dependence of the extracted sheet resistance of 2DES for AlO_x/Co/LaAlO₃/SrTiO₃(111) and Pt/LaAlO₃/SrTiO₃(111) for 3 ML and 6 ML of LaAlO₃ respectively. We discuss the possibility of separating out the contribution of the 2DES from the combined resistance in the supplementary part. In addition, in Figure S3 (b), we show the sheet resistance of a pristine LaAlO₃/SrTiO₃(111) film for 9 ML of LaAlO₃.

While suppression of t_c upon Co capping is observed for both the (111) and (001) LaAlO₃/SrTiO₃ interfaces, a reduction of t_c upon Pt capping occurs only for (111) interface, whereas an increase in t_c is found for the Pt capped (001) interface [31].

and 5 for the systems with Co and Pt capping. While for Co capping the internal electric field within LaAlO_3 is quenched, for Pt there is still a considerable internal electric field, albeit smaller than for the bare LaAlO_3 film. Thus the different critical thicknesses for Co and Pt can be rationalized by the the different size of p -type Schottky barriers that form between LaAlO_3 and the metal contact: 2.5 eV (Co) vs. 1 eV (Pt). In contrast for the (001)-oriented interface the p -type Schottky barriers were similar for a Co and Pt contact (~ 2.3 eV) [29], leading to similar band diagrams despite the difference in work function.

TABLE I. Calculated work functions (in eV) for different LaAlO_3 thicknesses (3, 6, and 9 ML), surface terminations (Al and LaO_3), partial surface hydrogenation [$\text{LaO}_2(\text{OH})$], different metal cappings (Pt and Co), and of free-standing Pt(111) and Co(111) slabs of 7 ML thickness unstrained and strained to the lateral lattice constant of SrTiO_3 .

	3 ML	6 ML	9 ML
Al	4.478	4.599	4.534
LaO_3	5.153	5.042	5.146
$\text{LaO}_2(\text{OH})$	4.186	4.193	4.631
LaO_3/Pt	6.080	6.076	6.072
LaO_3/Co	4.876	4.487	
	Pt(111)	Co(111)	
Free-standing	5.688	5.121	
Strained@ a_{STO}	5.734	4.780	

The right panels in Figs. 3, 4 and 5 also show the spatial distribution of the quasi 2DES within $\text{LaAlO}_3/\text{SrTiO}_3(111)$ with and without a metal capping layer, integrated from -0.2 to the Fermi level. In all cases, the Ti $3d$ bands in the SrTiO_3 part host the 2DEG. The characteristic shape of the electron clouds around the Ti positions indicate a predominant e'_g orbital polarization. We also note that for the systems covered by Pt or Co, a second conducting channel is present in the surface layer.

C. Metal capping effect on superconducting properties

The DFT+ U calculations presented above unveil the origin of the critical thickness for a metal-to-insulator transition in $\text{LaAlO}_3/\text{SrTiO}_3(111)$ and the role of metal capping. We now turn to study the effect of metal capping on superconductivity for the (001) and (111) interfaces. Surprisingly, we find that all the (111) samples, which show conductivity upon metal capping, also show superconductivity. In Tables 2, we summarize the properties of the (001) and (111) interfaces with various LaAlO_3 thickness and different metal capping. Fig. 6 displays the normalized resistance ($R/R_{(0.5K)}$) as a function of temperature for different (001) and (111) interfaces at the critical thickness t_c with Co and Pt capping.

To make sure that the observed superconductivity is a two-dimensional (2D) interfacial effect and not a spuri-

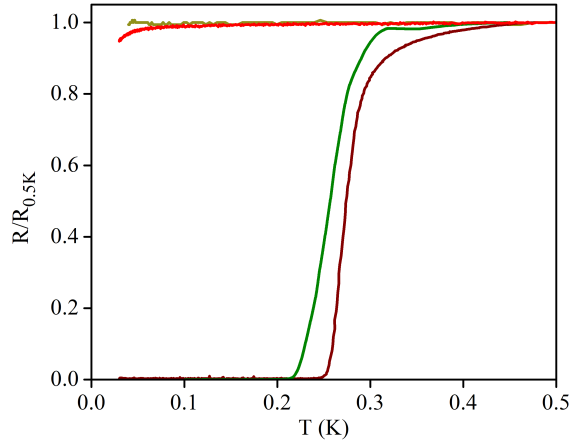


FIG. 6. The normalized resistance ($R/R_{(0.5K)}$) of $\text{AlO}_x/\text{Co}/\text{LaAlO}_3(3\text{ML})/\text{SrTiO}_3(001)$ (Dark Yellow), $\text{Pt}/\text{LaAlO}_3(9\text{ML})/\text{SrTiO}_3(001)$ (Red), $\text{AlO}_x/\text{Co}/\text{LaAlO}_3(3\text{ML})/\text{SrTiO}_3(111)$ (Olive), and $\text{Pt}/\text{LaAlO}_3(6\text{ML})/\text{SrTiO}_3(111)$ (Wine). The $\text{LaAlO}_3/\text{SrTiO}_3(111)$ interfaces capped with Co and Pt show a superconducting transition and the corresponding critical thickness of LaAlO_3 is 3 and 6 Monolayer respectively.

ous one resulting from the metal deposition process, we studied the temperature dependence of the perpendicular and parallel critical field. We show in the supplementary information that they both follow the expected 2D Ginzburg-Landau temperature dependence (See Figure S4 of supplemental information). The resulting coherence length (ξ) and the superconducting effective layer thickness (d) are displayed in Fig. S5.

In Fig. 7 we show the behavior of the critical temperature and critical fields as a function of gate voltage for Co and Pt capped (111) interface. The dome-shaped gate dependence and the values of T_c , the perpendicular (H_\perp) and the parallel critical field (H_\parallel) are similar to the bare $\text{LaAlO}_3/\text{SrTiO}_3(111)$ interface [17, 25].

One may claim that the absence of superconductivity in the Co-capped $\text{LaAlO}_3/\text{SrTiO}_3(001)$ is due to the Co ferromagnetism. Indeed, as shown previously in Fig. 1, the Co layer shows an anomalous Hall effect. However, for such a thin cobalt film, one expects the magnetic coupling to be short-ranged with a negligible effect on the 2DES.

How can we understand the robustness of superconductivity in (111) interfaces? Previous experimental studies on $\text{LaAlO}_3/\text{SrTiO}_3(111)$ interfaces show that even at strong negative gate voltages, superconductivity remains intact [25, 26]. On the theory side, the curvature of the Fermi contour changes quickly upon charge accumulation, and both the conducting and superconducting bands get an equal contribution from the three degenerate t_{2g} orbitals[17]. By contrast, for (001) interface, there is a distinct less mobile, non-superconducting band and

TABLE II. Summary of the different samples measured for various thickness of LaAlO_3 upon different metal capping and corresponding nature of the interface. Note: For (001) interface one monolayer corresponds to one unit-cell. "Yes" in the superconductivity column means that the sample reached zero resistance at some gate voltage and "Partial" means that a drop in resistance is observed at low temperatures in some of the gate voltages without reaching zero resistance

Interface	Metal layer	Thickness (ML)	Conducting	Super-Conducting
(111)	Co	1	No	No
(111)	Co	2	No	No
(111)	Co	3	Yes	Yes
(111)	Co	4	Yes	Yes
(111)	Co	6	Yes	Yes
(111)	Co	7	Yes	Yes
(111)	Pt	5	No	No
(111)	Pt	6	Yes	Yes
(111)	Pt	9	Yes	Yes
(111)	Pt	12	Yes	Yes
(001)	Co	2	Yes	No
(001)	Co	3	Yes	No
(001)	Pt	9	Yes	Partial
(001)	Ag	3	Yes	Partial
(111)	Bare	9	Yes	Yes
(001)	Bare	10	Yes	Yes

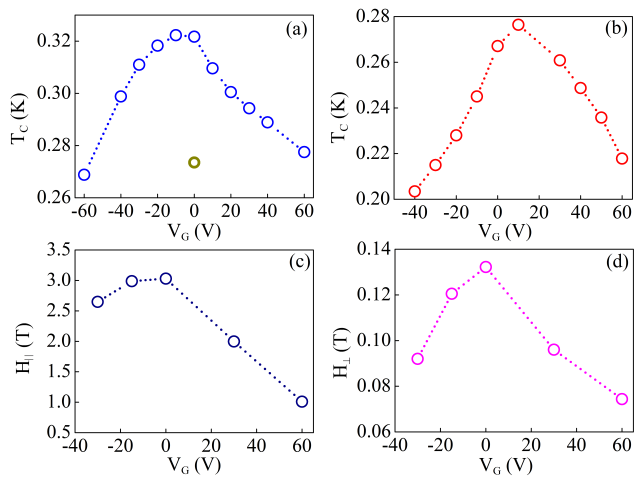


FIG. 7. (a) The critical temperature (T_c) of Pt/LaAlO₃(6ML)/SrTiO₃(111) interface, the dark yellow circle shows the T_c of the as cooled film. (b), (c) and (d) are superconducting critical temperature, perpendicular critical field and parallel critical field respectively of the AlO_x/Co/LaAlO₃(3ML)/SrTiO₃(001) interface as a function of gate voltage. The values of these critical parameters are in good agreement with that of the uncapped interface. Note: T_c is defined as a temperature where the value of resistance drops by 50% of its value at 0.5 K.

a mobile superconducting band due to the very different effective masses of the light d_{xy} and heavy d_{yz} , d_{xz} bands. It is possible that the inter-band repulsion [34] results in shifting the second band to higher energy leaving only the metallic state. Our DFT+ U calculations show that the e'_g bands lie lowest and contribute to conductivity

at the (111) interfaces suggesting that this type of band plays a significant role in the observed superconductivity.

IV. SUMMARY

Capping of Co and Pt on LaAlO₃/SrTiO₃(111) interface reduces the critical thickness for the onset of conductivity from 9 ML for the bare interface to 6 and 3 ML for platinum and cobalt respectively. DFT+ U calculations are consistent with the experimental observations. Importantly, all conducting (111) interfaces are also superconducting at low temperatures. This is in contrast with the (100) interfaces where conductivity emerges before superconductivity. The DFT+ U results suggest that the different behavior of the superconducting state for the (111) and (100) interfaces with metal capping is related to the different orbital contribution to the 2DES: superconductivity takes place in the e'_g for the (111) interface, whereas for the (100) interface, population of the heavy d_{xz} , d_{yz} bands is important for superconductivity.

Our findings suggest that proper choice of metal on top of the LaAlO₃ barrier can tune the barrier strength for various applications such as superconducting tunneling devices and ferromagnetic tunnel junction.

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