

Two-Dimensional Quantum Oscillations of the Conductance at LaAlO₃/SrTiO₃ Interfaces

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(Received 28 July 2010; published 1 December 2010)

We report on a study of magnetotransport in LaAlO₃/SrTiO₃ interfaces characterized by mobilities of the order of several thousands cm²/V s. We observe Shubnikov-de Haas oscillations whose period depends only on the perpendicular component of the magnetic field. This observation directly indicates the formation of a two-dimensional electron gas originating from quantum confinement at the interface. From the temperature dependence of the oscillation amplitude we extract an effective carrier mass $m^* \simeq 1.45m_e$. An electric field applied in the back-gate geometry increases the mobility, the carrier density, and the oscillation frequency.

DOI: 10.1103/PhysRevLett.105.236802

PACS numbers: 73.20.-r, 71.18.+y, 73.21.-b, 73.40.-c

Several experimental and theoretical studies have uncovered a number of remarkable electronic properties of the interface between the complex oxides LaAlO₃ and SrTiO₃ [1–5]. One of the main unresolved issues pertains to the dimensionality of the conducting layer [6]. While it is now clear that, using appropriate growth and annealing conditions, a confined metallic and superconducting electron gas can be formed at such interfaces [7,8], no conclusive demonstration of two-dimensional character in the normal state has been obtained so far.

X-ray absorption spectroscopy experiments [9], as well as density functional theory calculations [10], indicate that an orbital reconstruction occurs at this interface. These studies reveal that, even at room temperature, the degeneracy of the Ti 3d_{t_{2g}} levels is lifted and the first available states for the conducting electrons are generated from 3d_{xy} orbitals, which give rise to strongly two-dimensional bands and present a negligible interplane coupling. Moreover, the momentum quantization in the quantum well brings about a subbands fine structure which was calculated [10,11] but not yet confirmed experimentally. The presence of a very strong spin-orbit coupling adds further complexity to the low-energy electronic structure [12,13].

The Fermi surface of two-dimensional electronic states generates clear experimental signatures in the Shubnikov-de Haas (SdH) effect: for a two-dimensional electron gas (2DEG) the quantum oscillations depend only on the perpendicular component of the magnetic field. Previous studies reported quantum oscillations in LaAlO₃/SrTiO_{3-δ} heterostructures characterized by a large carrier density (of the order of 10¹⁶ cm⁻²) delocalized in the SrTiO₃ substrate [1,14]. The lack of dependence of the oscillations on the field orientation points to a three-dimensional Fermi surface consistently with previous studies in Nb-doped SrTiO₃ single crystals [15]. Very recently, quantum oscillations with two-dimensional character have been reported

for delta-doped SrTiO₃ [16,17]. However, in LaAlO₃/SrTiO₃ heterostructures where the electrons are confined at the interface [6], magnetotransport studies have been carried out so far in the diffusive regime, where the scattering time is not sufficiently long to give rise to well defined Landau levels [12,18–20]. These transport studies—together with spectroscopic investigations and theoretical analyses mentioned above [5,9,11]—have highlighted important differences between doped SrTiO₃ and the LaAlO₃/SrTiO₃ interface system.

The requirements for observing quantum conductance oscillations are [21]

$$\omega_c \tau > 1, \quad (1)$$

$$\hbar \omega_c > k_B T, \quad (2)$$

where $\omega_c = eB/m^*$ is the cyclotron frequency, m^* is the carrier effective mass, e the elementary charge, B the applied magnetic field, T is the temperature, and τ is the transport elastic scattering time. To fulfill these conditions with temperatures of the order of 1 K and magnetic fields of a few Tesla, mobilities of 10³ cm²/V s or more are required. Existing strategies to reach this value rely on the use of the electrostatic field effect [4,22] or defect control [16,23] to improve the quality of the devices. However, currently available samples have not yet allowed us to access the regime where quantum conductance oscillations are unambiguously visible.

In this Letter we present a magnetotransport study performed in LaAlO₃/SrTiO₃ interfaces in which the mobility has been boosted by an optimization of the growth conditions, reaching the unprecedented value of 6600 cm²/V s. In these samples we observe quantum oscillations in the electrical resistance as a function of magnetic field whose frequency depends only on the perpendicular component of the magnetic field. An electric

field applied to the back-gate modulates the oscillation frequency. These results demonstrate the presence of two-dimensional electronic states originating from quantum confinement at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface.

We fabricated high-mobility samples by depositing epitaxial thin films of LaAlO_3 on top of TiO_2 -terminated SrTiO_3 single crystals. The films were grown by pulsed laser deposition at $\sim 650^\circ\text{C}$ in $\sim 1 \times 10^{-4}$ mbar of O_2 with a repetition rate of 1 Hz. The fluence of the laser pulses was 0.6 J/cm^2 . The films growth was monitored *in situ* using reflection high energy electron diffraction (RHEED). After growth, each sample was annealed in 200 mbar of O_2 at about 530°C for 1 h and cooled to room temperature in the same oxygen pressure. The reduced growth temperature, with respect to standard deposition conditions ($\sim 800^\circ\text{C}$), significantly improves the crystalline quality of the films, as observed by x-ray diffraction and RHEED. The samples are metallic and superconducting below 100 mK (the analysis of the superconducting properties in this cleaner interface is certainly interesting, but goes beyond the scope of this manuscript and will be discussed elsewhere). They are characterized by mobility values (of the order of $5000 \text{ cm}^2/\text{Vs}$ at 1.5 K) that are approximately 1 order of magnitude higher than those observed in heterostructures grown with the deposition conditions used in the past [24]. The influence of the deposition conditions on the electron mobility is discussed in the supplementary material [25].

We have measured a total of 10 heterostructures with a thickness of the LaAlO_3 top layer between 5 and 10 unit cells, all of which exhibited clear quantum conductance oscillations. Some differences are present in the behavior of the oscillations observed in different samples. In particular, the amplitude (and precise shape) of the oscillations varies from sample to sample. In a few samples oscillations are visible directly in the raw magnetoresistance data; in most other samples it is useful to take the derivative of the resistance with respect to the magnetic field—or to remove a background—to make the oscillations fully apparent. Nevertheless, the overall phenomenology that we discuss here is common to the whole set of devices.

Figure 1(a) shows the variation of sheet resistance $\Delta R = R(B) - R(0)$ in response to the application of a magnetic field oriented perpendicular to the interface, recorded at different temperatures. The resistance measurements have been performed using a 4 point ac technique, with a current between 10 and 100 nA, in a standard Hall bar defined by photolithography, with a channel width of $500 \mu\text{m}$ and with voltage probes 1 mm apart [4]. Below $T = 7 \text{ K}$, oscillations superimposed on a positive background are observed for fields larger than 3 T (with $\mu = 5000 \text{ cm}^2/\text{Vs}$, $\omega_c\tau > 1$ for $B > 2 \text{ T}$; at $B = 8 \text{ T}$, $\omega_c\tau = 4$). The nonoscillatory background is consistent with previous magnetotransport studies of this system [12,19,20]. The numerical derivative of the resistance with respect to

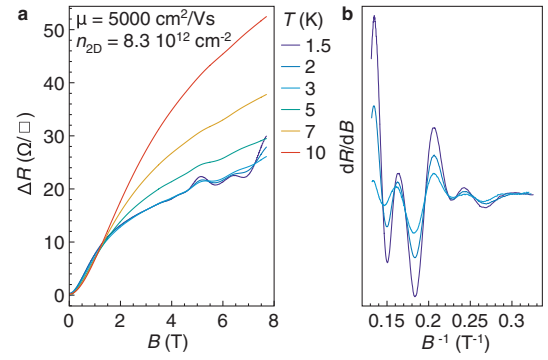


FIG. 1 (color). Shubnikov-de Haas oscillations of the $\text{LaAlO}_3/\text{SrTiO}_3$ interface. (a) Variation of resistance $\Delta R = R(B) - R(0)$ in response to the application of a magnetic field B oriented perpendicular to the $\text{LaAlO}_3/\text{SrTiO}_3$ interface, recorded at different temperatures T . (b) Numerical derivative dR/dB as a function of the inverse of the magnetic field.

magnetic field, presented in Fig. 1(b), reveals that the oscillations are periodic in $1/B$.

The dimensionality of the electronic states can be assessed by examining the angular dependence of the quantum oscillations. Figure 2(a) displays $R(B)$ measured at $T = 1.5 \text{ K}$ on a different sample, for different angles θ . The angle θ measures the inclination of the magnetic field with respect to the normal to the interface at a fixed azimuthal angle. At $\theta = 0^\circ$ the magnetic field is applied perpendicular to the interface, while for $\theta = 90^\circ$ the magnetic field vector lies in the plane of the interface, parallel to the current. At $\theta = 90^\circ$ we observe, consistently with previous reports, a fairly large negative magnetoresistance [12,19]. Figures 2(b) and 2(c) show the derivative of the

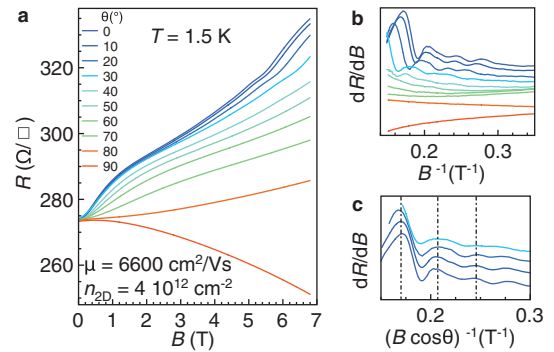


FIG. 2 (color). Angular dependence of the quantum oscillations. (a) Sheet resistance R as a function of magnetic field B recorded at different orientations (measured by the angle θ) with respect to the direction normal to the substrate. (b) Numerical derivative dR/dB as a function of the inverse of the magnetic field recorded at different orientations. (c) Numerical derivative dR/dB as a function of the inverse of the component of the magnetic field perpendicular to the plane of the interface. An offset has been introduced in each curve for clarity. The lines are a guide to the eye.

data as a function of B^{-1} and $(B \cos \theta)^{-1}$, respectively. It is apparent that the oscillation depends only on the perpendicular component of the magnetic field. This observation directly indicates that the oscillations arise from closed orbits in momentum space along a two-dimensional Fermi surface.

SdH oscillations represent a direct measurement of the area of the Fermi surface. Indeed, the Onsager relation $\omega = (\phi_0/2\pi^2)A$ establishes a direct proportionality between the cross-sectional area of the Fermi surface normal to the magnetic field A and the frequency of the oscillation ω (ϕ_0 is the magnetic flux quantum) [26]. Figure 3(b) displays the Fourier transform of the oscillatory component of the magnetoresistance of a sample characterized at 250 mK by a mobility of $2860 \text{ cm}^2/\text{Vs}$ and a Hall carrier density $n_{2D} = 1.05 \times 10^{13} \text{ cm}^{-2}$. A main broad peak is clearly visible in the Fourier transform at 35 T with a shoulder at 50 T. In other samples the shoulder appears as a small secondary peak. The main peak leads to an area of 0.33 nm^{-2} , which is only 0.1% of the Brillouin zone. Assuming a circular section of the Fermi surface, we can estimate the carrier density as $n_{2D} = \omega \nu_v \nu_s e/h$, where ν_v and ν_s indicate the valley and spin degeneracy, respectively. By taking $\nu_s = 2$ and a single valley we find $n_{2D} = 1.69 \times 10^{12} \text{ cm}^{-2}$ (the feature at 50 T would give an additional contribution of $2.4 \times 10^{12} \text{ cm}^{-2}$). The corresponding Fermi wavelength is $\lambda_F = 2\pi/k_F = 19 \text{ nm}$. This value is larger than the thickness of the electron gas at low temperatures ($\approx 10 \text{ nm}$) estimated in low-mobility samples using the anisotropy of the superconducting critical fields [8], infrared ellipsometry [27], and atomic force microscopy [6,7]. Apart from the apparent discrepancy between the density values extracted from the Hall effect measurements and the SdH oscillations (which will be discussed later), these findings are consistent with a picture

of a two-dimensional electron gas formed at the $\text{LaAlO}_3/\text{SrTiO}_3$ interface.

We now turn our attention to the temperature dependence of the oscillations amplitude, which contains important information regarding the carrier effective mass and the level of disorder. For this analysis we performed magnetoresistance measurements up to 15 T and down to 250 mK. The amplitude of the quantum oscillation, extracted from these measurements by subtracting a polynomial background [see Fig. 3(a)], decreases with increasing temperature.

The oscillations amplitude ΔR as a function of temperature T can be described by the relation [21]

$$\Delta R(T) = 4R_0 e^{-\alpha T_D} \alpha T / \sinh(\alpha T) \quad (3)$$

where $\alpha = 2\pi^2 k_B / \hbar \omega_c$, R_0 is the nonoscillatory component of the square resistance, and T_D is the Dingle temperature, which is a measure of the level of disorder. The best fit to Eq. (3) of our experimental data is shown in Fig. 3(c), for the largest maximum of the oscillations (the second largest gives the same result). The fitting parameters are the carrier effective mass (which enters the cyclotron frequency) and the Dingle temperature. We observe a good agreement between theory and experiment with $m^* = 1.45 \pm 0.02 m_e$ and $T_D = 6 \text{ K}$. Our estimation of the effective mass is similar to what has been observed in doped SrTiO_3 by quantum oscillations in thin films [16] and by optical conductivity in crystals [28]. We note also that in samples with an order of magnitude larger carrier density, infrared ellipsometry gives an effective mass that is a factor of 2 larger than what we find [27]. Finally, the broadening of the Landau levels that we find, determined by $k_B T_D \sim 0.5 \text{ meV}$, is not much smaller than their energy separation ($\hbar \omega_c \sim 1 \text{ meV}$ at 10 T). This explains the small amplitude of the oscillations that we observe.

On some of our samples we have also looked at the evolution of the quantum oscillations in devices equipped with a gate electrode, which consists of a metal layer deposited on the back side of the substrate (0.5 mm thick). Figures 4(a) and 4(b) display $R(B)$ measurements and $dR/dB(B^{-1})$ recorded at $T = 250 \text{ mK}$ and different gate voltages V . In conventional two-dimensional electron gases, the main effect of tuning the gate voltage is to modulate the carrier density, leading to a change in the Fermi surface, and thereby in the SdH oscillation frequency. Indeed, as expected, we observe a clear shift of the main peak of the oscillations [see Fig. 4(c)], whose frequency increases linearly with carrier density measured by Hall effect [29]. In performing these experiments, we also noted that in most devices the oscillations become more apparent at larger gate voltages. This is a consequence of the fact that the mobility increases with V [22] (in the device whose data are shown in Fig. 4 the mobility increases from $2400 \text{ cm}^2/\text{Vs}$ at 10 V to $3600 \text{ cm}^2/\text{Vs}$ at 120 V).

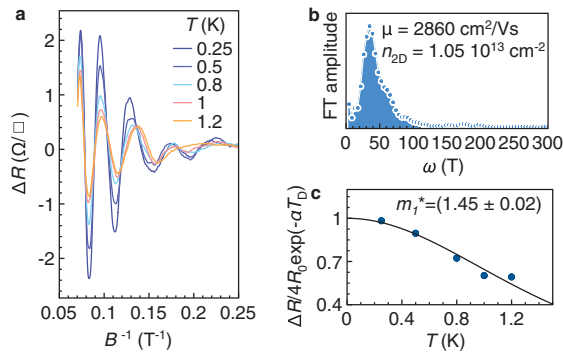


FIG. 3 (color). Temperature dependence of the Shubnikov-de Haas oscillations. (a) Oscillatory component of the sheet resistance ΔR as a function of the inverse of the magnetic field oriented perpendicular to the $\text{LaAlO}_3/\text{SrTiO}_3$ interface, recorded at different temperatures T . (b) Fourier spectrum of the resistance oscillation measured at 250 mK. (c) Blue dots: temperature dependence of the amplitude of the oscillation at 13.5 T. Solid line: best fit to Eq. (3) of the experimental data.

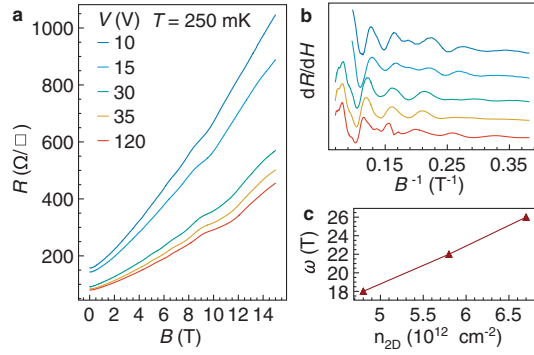


FIG. 4 (color). Field effect modulation of the Shubnikov–de Haas oscillations. (a) Sheet resistance R as a function of magnetic field B recorded at different gate voltages V at $T = 250$ mK. (b) Numerical derivative dR/dB as a function of the inverse of the magnetic field showing the effect of the gate voltage on the resistance oscillations. (c) Main frequency of the oscillation ω as a function of carrier density n_{2D} .

We conclude that the realization of high-quality $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces enables us to reveal the presence of two-dimensional electronic states. One issue that remains to be solved is the apparent mismatch between the values of the carrier density estimated from the Hall effect and from the SdH oscillations (approximately a factor of 4–5 if we consider only the main peak, a factor of 2 if we consider also the peak at 50 T). This discrepancy could be due to a nonoscillatory parallel transport channel whose origin remains to be determined, but other possibilities can also be envisioned. For instance, in the experiments the limited number of periods observed may not be sufficient to resolve all peaks in the spectrum of the SdH oscillations. Also, even though band-structure calculations predict the presence of only one valley [10] in the conduction band of $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces, it cannot be excluded that in reality multiple valleys are present. Finally, effects associated to the spin of the electrons may be important. On the one hand, a strong spin-orbit interaction is known to affect SdH oscillations [12,30]. On the other hand, since the effective mass is larger than the free electron mass, the Zeeman splitting is larger than the Landau level spacing even when the gyromagnetic factor is $g = 2$, which brings the system in an unconventional regime. Additional studies are required to elucidate this point, which most likely will require samples of even higher quality and higher magnetic fields.

The observation of two-dimensional transport originating from quantum confinement in $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces establishes a connection between this system and semiconducting heterostructures based on III-V compounds. In contrast to these more conventional systems 2DEGs at $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces are characterized by very strong spin-orbit interaction, much lower Fermi energy, higher effective mass, not to mention the occurrence of superconductivity. The availability of high-quality

$\text{LaAlO}_3/\text{SrTiO}_3$ interfaces gives access to two-dimensional electron physics in an entirely unexplored parameter regime, which is likely to disclose new phenomena not yet observed in the diffusive transport limit that has been investigated so far.

We thank T. Giamarchi, D. Jaccard, and D. van der Marel for useful discussions and Marco Lopes for his technical assistance. We acknowledge financial support by the SNSF through the NCCR “Materials with Novel Electronic Properties” MaNEP and Division II, by the EU through the projects Nanoxide and OxIDes.

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