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Two-dimensional electron gas in δ-doped $SrTiO_3$

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It is shown that Shubnikov-de Haas oscillations in $SrTiO_3$ δ-doped with La can be understood as arising from a two-dimensional electron gas of one subband immersed in the space charge layer. Despite the inherent complexity of a subband that is derived from four d-band states near the conduction-band minimum of $SrTiO_3$, the quantum oscillations can be modeled quantitatively by recognizing that the magnetic field (B) induced effective spin splitting and Landau-level splitting are comparable. The oscillations are not strictly periodic in $1/B$, which can be understood as caused by a weak dependence of the electron density on the magnetic field in the subband that produces the observed oscillations.

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Understanding two-dimensional quantum phenomena in $SrTiO_3$ is important both from a fundamental physics perspective and for device applications. For example, confined electron gases in $SrTiO_3$ have attracted attention for their potential of combining unique field-tunable, magnetic, and superconducting phenomena.[1–4] The *conduction band* of $SrTiO_3$ consists of a heavy and a light electron band and a spin-orbit split-off band,[5,6] resembling in many ways the *valence band* of the much more widely studied conventional semiconductors. Two-dimensional *hole* gases in III-V heterostructures have been extensively characterized because of the unique spin and charge phenomena that derive from the valence-band structure and which are of interest for novel devices.[7–9] In addition, two-dimensional electron gases in transition-metal oxide heterostructures open the possibility of other novel physical phenomena because the low-dimensional transport occurs in d-electron bands, which are subject to strong electron-electron interactions known to produce Mott insulators. Although several different approaches toward two-dimensional electron gases in $SrTiO_3$ have been explored in the literature, so far only δ-doped layers have exhibited sufficient electron mobilities to allow for the observation of quantum oscillations with two-dimensional character.[10]

Similar to the development of III-V semiconductor heterostructures, reduction in the intrinsic defect concentrations in $SrTiO_3$ films allows for improvements in the low-temperature electron mobility, crucial for the study and interpretation of quantum phenomena. For example, we have recently shown that $SrTiO_3$ thin films grown by molecular-beam epitaxy (MBE) exhibit electron mobilities that are far greater than those in thin films grown by other methods and even exceed those of $SrTiO_3$ single crystals.[11]

The goal of this Rapid Communication is to communicate an understanding of the magnetotransport in MBE-grown δ-doped $SrTiO_3$ films. We show that only the electrons in one of the subbands confined by the δ-doping potential have sufficient mobility to exhibit two-dimensional Shubnikov-de Haas oscillations. Guided by the similarity of the confined d-band electron states in $SrTiO_3$ to the confined hole systems in conventional semiconductors, we interpret the quantum oscillations in terms of spin and Landau-level splitting of a single nonparabolic band derived from the manifold of states at the $SrTiO_3$ conduction band minimum.

δ-doped epitaxial $SrTiO_3$ films were grown at a substrate temperature of 870 °C on (001) $SrTiO_3$ single crystals by MBE. Details of the growth process, the structural characteristics, and the electrical properties of uniformly doped films, including their electron mobilities, have been reported in detail elsewhere.[11–13] All substrates and undoped films were insulating.[11] The δ-doped structure consisted of a 300-nm-thick buffer layer of undoped $SrTiO_3$, followed by a ~3-nm-thick La-doped $SrTiO_3$ layer, which was capped with 70 nm of undoped $SrTiO_3$ to reduce the effects of surface depletion. La concentration depth profiles obtained using secondary-ion-mass spectrometry (SIMS) showed that the doping concentration profiles were abrupt, indicating no detectable diffusion of La out of the δ-doped layer.[14] Low-temperature magnetotransport measurements were made in Hall bar geometry with rectangular-shaped samples (10×4 mm^2) with Ohmic contacts [Al(40 nm)/Ni(20 nm)/Au(150 nm)] deposited by electron-beam evaporation through a shadow mask. Measurements were carried out in a physical property measurement system (PPMS, Quantum Design), which allowed for measurements down to 400 mK using a He^3 cryostat and for sample rotation up to 90° at temperatures above 1.8 K.

Longitudinal and transverse magnetoresistance was measured from 0 to 14 T. The overall longitudinal magnetoresistance (ΔR_{xx}) was positive and increased by ~15% at 14 T; the transverse magnetoresistance was weakly nonlinear. Both features indicate transport by two or more sets of carriers with different mobilities, consistent with more than one occupied subband (see below). Hall measurements were carried out using 10 μA/17 Hz currents and sweeping the magnetic field (B) between ±1 T. The low-field (±1 T) magnetoresistance corresponded to a two-dimensional electron density of 3×10^{13} cm^{-2} at 1.8 K and Hall mobility of 1500 cm^2/V s.[14] ΔR_{xx} vs B was recorded with a 1 kHz/50 μA current and lock-in amplifier, from 0 to 14 T, at temperatures from 0.41 to 3.0 K. At the lowest temperatures weak Shubnikov-de Haas oscillations appear in the

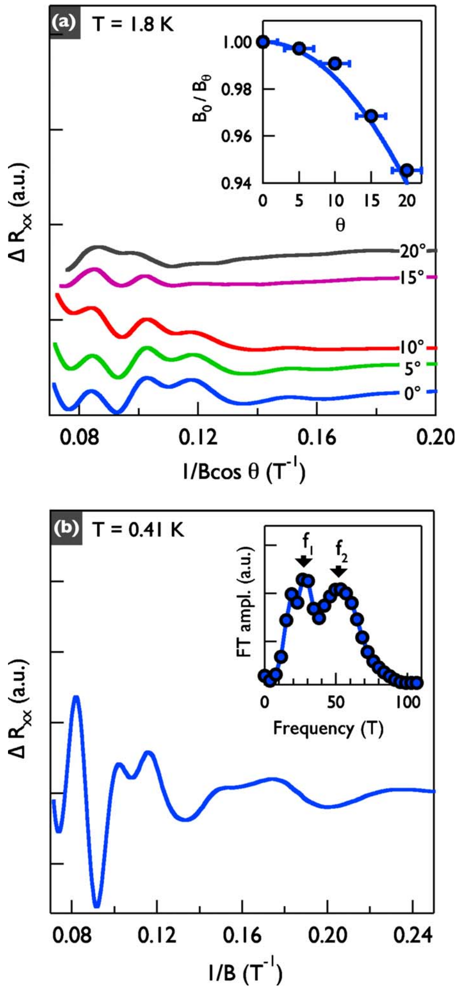

FIG. 1. (Color online) (a) Shubnikov-de Haas oscillations at $T=1.8$ K as a function of the inverse of the perpendicular component of the magnetic field. The inset shows the θ dependence of a Shubnikov-de Haas oscillation minimum position in a magnetoresistance vs $1/B$ plot (relative to its position at $\theta=0°$), confirming the two-dimensional electron gas behavior. For comparison, the solid line shows the ideal cos θ dependence. (b) Shubnikov-de Haas oscillations as a function of the inverse of the magnetic field at $\theta=0°$ and $T=0.41$ K. The inset shows the Fourier transform of the magnetoresistance data, showing two harmonically related frequencies (27 and 54 T).

magnetoresistance. To recover the oscillating component of ΔR_{xx} the data was smoothed and a background in the form of an arbitrary cubic polynomial was subtracted.[14]

Figure 1(a) shows the Shubnikov-de Haas oscillations measured at 1.8 K as a function of the inverse of the surface-normal component of the magnetic field, $1/B$ cos θ, for different tilt angles θ between B and the sample normal. For an ideal two-dimensional system, the Landau-level splitting and the cyclotron frequency ω_c depend only on B cos θ and the minima (maxima) in ΔR_{xx} should thus appear at the same $1/B$ cos θ value, as is indeed observed. The inset shows the relative (to the position at $\theta=0°$) position of one of the minima in a magnetoresistance vs $1/B$ plot as a function of θ, showing good agreement with the expected cos θ dependence (shown as solid line).

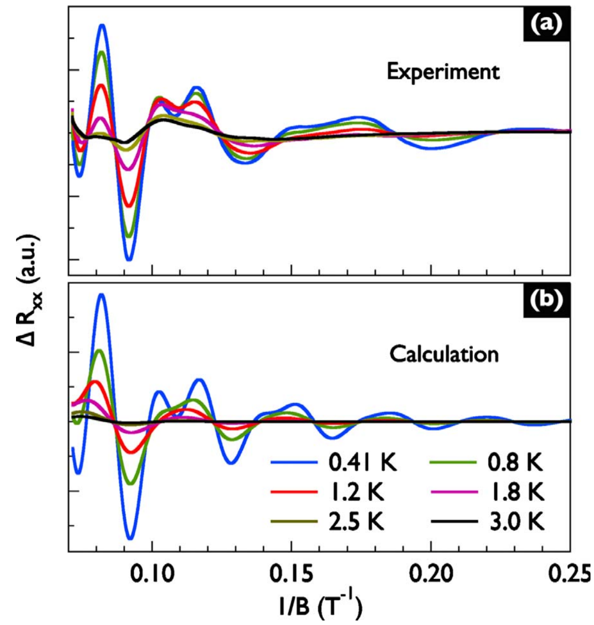

FIG. 2. (Color online) (a) Experimental Shubnikov-de Haas oscillations as a function of the inverse of the magnetic field at temperatures between 0.41 and 3 K. The amplitude of the oscillations decreases with increasing temperature. (b) Calculated Shubnikov-de Haas oscillations at temperatures between 0.41 and 3 K using a single, spin-split subband model as described by Eq. (1). The values for the parameters in Eq. (1) were obtained by fitting to the low-temperature data, using R_0, g, T_D, and $\Delta 1/B$ as fit parameters. The effective mass used in the fits was determined from the experimental temperature dependence.

Figure 1(b) shows the Shubnikov-de Haas oscillations as a function of $1/B$ at $\theta=0°$, measured at a temperature of 0.41 K. It is apparent that there are two dominant frequencies or periods, causing weak minima to appear between the strong minima. Indeed, a Fourier transform [shown as inset in Fig. 1(b)] exhibits two broad peaks, at 27 and 54 T, which appear to be harmonically related. Figure 2(a) shows the Shubnikov-de Haas oscillations as a function of temperature. The low-frequency (27 T) oscillation in $1/B$ is most apparent at elevated temperatures while the high-frequency oscillation tends to express itself at low temperatures.

Similar Shubnikov-de Haas oscillations for Nb-δ-doped $SrTiO_3$ have been interpreted as arising from three subbands.[10] Given the two frequencies in the Fourier transform, the data shown in Fig. 1(b) was fit to a two subband model, using the standard equation for a two-dimensional electron gas without spin splitting.[14,15] The two-subband model was able to reproduce the high-field features but only for frequencies (21 and 54 T) that do not match those in the experiments.[14] The low-field data could not be matched, even qualitatively.[14]

Given an electron mass of $SrTiO_3$ of around $1-2m_e$ (Ref. 6) and a Landé g factor of ~2, the Landau and spin splittings should be roughly comparable. Thus a model with Landau splitting alone, assuming $g=0$, as used in Ref. 10, may be too constrained. We therefore next discuss the possibility that the relatively complex Shubnikov-de Haas oscillations can be understood as arising from the two-dimensional electron gas in a single subband but with a Landau-level spectrum that includes spin splitting. We use the three-dimensional expression[16] given by Roth and Argyres[17,18] to describe the Shubnikov-de Haas oscillations because it includes the effects of spin splitting and is applicable[16] to weak sinusoidal oscillations due to strong scattering,

$$\frac{\Delta R_{xx}}{R_0} = \frac{5}{2}\sum_{s=1}^{\infty} b_s \cos\left(\frac{2\pi E_F}{\hbar\omega_c}s - \frac{\pi}{4}\right), \tag{1a}$$

where $\hbar$ is the reduced Planck's constant, E_F the Fermi energy, ω_c the cyclotron frequency, $\omega_c = eB/m^*$, where m^* is the effective mass. Translating to the two-dimensional case, $\frac{2\pi E_F}{\hbar\omega_c}$ is replaced by $\frac{2\pi nh}{2eB}$, where n is the electron density (the factor of 2 accounts for spin degeneracy). The amplitude, b_s, is given by[17,18]

$$b_s = \frac{(-1)^s}{\sqrt{s}}\left(\frac{\hbar\omega_c}{2E_F}\right)^{1/2}\frac{2\pi^2 s k_B T/\hbar\omega_c}{\sinh(2\pi^2 s k_B T/\hbar\omega_c)} \times\exp\left(-\frac{2\pi^2 s k_B T_D}{\hbar\omega_c}\right)\cos\left(\frac{\pi s g m^*}{2m_e}\right), \tag{1b}$$

where k_B is the Boltzmann constant, T_D the Dingle temperature, and m_e is the free-electron mass. The relative strength of the weak and strong minima is determined by the gm^* product, the periodicity by n, and the temperature dependence is determined by m^*. The low-temperature (0.41 K) oscillations were fit to Eq. (1), including first and second harmonics ($s=1,2$) and with R_0, T_D, g, and the fundamental period $\Delta 1/B$ ($\Delta 1/B = 2e/nh$) as the fit parameters. The effective mass m^* was obtained from the temperature dependence of the relative amplitude of the two strongest oscillations as a function of temperature.[14,15] The value obtained for m^* is $\sim 1.56 \pm 0.08 m_e$.[14] This mass is reasonable for states near the conduction-band minimum of $SrTiO_3$.[5,6] The parameters obtained from the fit of the experimental data to Eq. (1) were $T_D = 0.53$ K and $(\Delta 1/B)^{-1} = 27$ T, the same frequency as obtained from the Fourier transform in Fig. 1(b). Because of the periodicity of $\cos(\frac{\pi s g m^*}{2m_e})$, the fits produces a series of possible g factors, 0.698, 1.86, 3.25, etc. It is clear that spin splitting is important but the g factor should be determined by additional experiments. Next, the temperature dependence was calculated with these values, as shown in Fig. 2(b). Comparison with the experiments shown in Fig. 2(a) shows that in the low-$1/B$ (high-field) region a quantitative match is achieved with regards to the temperature dependence, the periodicity, and the relative strengths of the harmonics. The quantum scattering time is $\tau_q = \hbar/2\pi k_B T_D = 2.29 \times 10^{-12}$ s, corresponding to a quantum mobility of 2600 $cm^2\ V^{-1}\ s^{-1}$, which places a lower limit on the transport mobility for this subband. The observed frequency corresponds to a carrier concentration of 1.3×10^{12} cm^{-2}. Thus, similar to what was observed in Ref. 10, the electron density in the subband with sufficient mobility to exhibit Shubnikov-de Haas oscillations is only 4% of the total Hall density. While a quantitative model of the subband states in the δ doping potential of $SrTiO_3$ does not yet exist, these states should be qualitatively similar to those found in conventional semiconductors.[19] The lowest energy state is tightly bound, has the largest two-dimensional density, is least screened, and experiences the greatest scattering from the dopant ions (lowest mobility). Conversely the highest bound states will have the largest spatial extension, have fewer two-dimensional electrons, and suffer the least scattering from the dopants. It is reasonable to assign the Shubnikov-de Haas oscillations, observed and modeled here, to the highest subband. In principle, other subbands could add to the Shubnikov-de Haas oscillations but with different periods and amplitudes. We conclude from their absence that their mobilities are too low to be observed.

In the above discussion we have focused on the low-$1/B$ (high-B) region. Comparing the calculations with the experiment over the entire $1/B$ range shows that although weak and strong features in the oscillations and their temperature dependence agree qualitatively with the model also in the high-$1/B$ (low-B) region, the frequency appears to shift (note the expanding separation in $1/B$ in the experiment). This behavior is contrary to the usual simple periodicity in $1/B$ of a two-dimensional electron gas.

The rigorous periodicity in $1/B$ of the oscillations of a two-dimensional electron gas arises from the quantization of the number of states that can be accommodated in a single spin Landau level. The quantization is independent of the complexity of the system, such as the nonparabolicity of bands. A level is completely occupied if it has an electron density eB/h. If the two-dimensional electron density is fixed then a $1/B$ periodicity follows invariably. The apparent departure from $1/B$ periodic behavior despite reproducing the essential structure in the Landau/spin spectrum implies *that the density is not fixed for this electric subband and varies slightly in the strong magnetic field*. Since this subband is in equilibrium with a much larger number of electrons in the δ-doping potential, it can also exchange carriers with the much larger bath of electrons and thus, *a priori*, there is no reason to assume that the density cannot change.

Furthermore, the manifold of bands at the bottom of the conduction band in $SrTiO_3$ does not have a simple parabolic dispersion;[5] the Landau-spin spectrum in a quantizing magnetic field will not expand as a simple, linear Landau fan diagram. The analogy with the two-dimensional hole Landau spectrum in III-V semiconductors is very instructive here.[20,21] Nonlinearities arise from coupling as a result of the interplay of the magnetic field, the confining potential of the δ-doped layer, and the tetragonal distortion in the $SrTiO_3$ at low temperatures.[20,21] Assuming a constant mass for the entire range of magnetic fields is an oversimplification. The complexities described by nonparabolicity do not, however, change the need to invoke a change in density in this subband with magnetic field. Future theoretical work should be directed toward modeling and understanding of the subband structure of confined electrons in $SrTiO_3$.

In conclusion, we have observed Shubnikov-de Haas oscillations in the magnetotransport of electrons in La-δ-doped $SrTiO_3$. The two-dimensional quantum oscillations appear to arise from a subband that has a small fraction of the total carrier density. The complex pattern and its temperature dependence can be understood in a quantitative manner by recognizing that the spin splitting and Landau-level splitting are of the same order of magnitude and we extract a measure of g and m^*. A departure from strict $1/B$ periodicity indicates a magnetic field dependence of the electron density in this subband. Finally, the results presented here point toward strong spin-related effects in two-dimensional electron gases in $SrTiO_3$.

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