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High-spin polarized Co-doped (La,Sr)TiO$_3$ thin films on high-mobility SrTiO$_3$ substrates

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Abstract

We report on the magnetotransport properties and spin polarization of Co-doped (La,Sr)TiO$_3$ (Co-LSTO) thin films in which the host oxide is a strongly correlated metal with a high density of carriers. In order to probe the spin polarization, we have performed tunneling magnetoresistance (TMR) measurements on magnetic tunnel junctions associating Co-LSTO and Co electrodes. A large spin polarization of around −80% has been determined at low temperatures, indicating that the carriers in this system are highly spin-polarized. We also report on the planar magnetotransport experiments on Co-LSTO films grown on SrTiO$_3$ substrates. The electrical properties of these samples are strongly dependent on the growth conditions and large electronic mobilities as high as $10^4\,\mathrm{cm^2/V\,s}$ at $T < 10\,\mathrm{K}$ are observed for specimens grown at the lowest oxygen pressure. We show that this high-mobility state is three-dimensional and we relate it to the doping of the SrTiO$_3$ substrate with oxygen vacancies.

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Keywords: Spintronics; Spin injection; Shubnikov–de Haas oscillations; Mobility

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The research on magnetic semiconductors is one of the most relevant subjects of the nowadays spintronics. The discovery of room temperature ferromagnetism in Co-doped TiO$_2$ [1] has triggered an intense research on other diluted systems, but the origin of ferromagnetism is not still well understood. Most of these works have been aimed at the ferromagnetic properties, although a spin polarization of carriers is an essential requirement for the use of DMOS in spintronics. On the other hand, further developments in spintronics require the combination of ferromagnets with semiconductors in order to, for instance, modulate spin-polarized transport by a gate voltage [2]. One of the best solutions for that purpose would be the realization of the heterostructures that combine highly spin-polarized ferromagnets and nonmagnetic materials with similar resistivities [3], to enable efficient spin injection in the ohmic regime and low carrier density, to enable gate voltage effects.

Here, we report on the magnetotransport properties and spin polarization of (La,Sr)Ti$_{1-x}$Co$_x$O$_3$ (Co-LSTO) thin films grown on SrTiO$_3$ (STO) substrates in low oxygen pressure conditions. We have demonstrated, through spin-dependent tunnel experiments, a high spin polarization of Co-LSTO at low temperatures. Furthermore, we have found that the conditions required to grow the diluted magnetic system with such large spin polarization also generate a high-mobility state in the STO substrate. As discussed later, the high-mobility state is created in STO over the thickness, which is of the order of hundreds of microns.

Co-LSTO and LSTO epitaxial thin films were grown on STO (0 0 1) or LAO (0 0 1) (LAO:LaAlO$_3$) substrates

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by pulsed laser deposition (PLD) from (La,Sr)Ti$_{1-x}$Co$_x$O$_3$ targets with $x = 0.02$ and $x = 0$ [4]. The La/Sr ratio was about 2:1 and the deposition oxygen pressure (P_{O_2}) was varied between 6×10^{-7} and 10^{-4} mbar.

As a direct way to probe the spin polarization of Co-LSTO, we have performed transport measurements in Co-LSTO-based magnetic tunnel junctions. First, a 150 nm thick Co-LSTO film was deposited as a bottom electrode, which was followed by a 2.8 nm LAO barrier on a STO substrate; both were grown at an oxygen pressure $P_{O_2} = 6 \times 10^{-7}$ mbar. Such Co-LSTO/LAO bilayers were subsequently covered by ex situ sputtering of a Co/CoO/Au stack, processed into tunnel junctions and measured in a standard 4-wire DC configuration.

The main result of the tunneling magnetoresistance (TMR) measurements on these magnetic tunnel junctions is shown in Fig. 1. From the large (almost 20%) low-temperature TMR, a large spin polarization (of the order of −80%) has been inferred [5], indicating that the carriers in this system are highly spin polarized. Our previous extensive structural analysis of Co-LSTO, combining high-resolution transmission electron microscopy, Auger electron spectroscopy and X-ray magnetic circular dichroism, excluded the presence of any parasitic phase (e.g. Co clusters, (La,Sr)CoO$_3$, etc.) in the Co-LSTO layer. We can therefore conclude that the observed high spin polarization is an intrinsic property of this system.

The current-in-plane magnetotransport measurements (resistance, magnetoresistance, and Hall effect) were performed on the Co-LSTO and LSTO single films grown at low oxygen pressure ($P_{O_2} \leqslant 10^{-6}$ mbar) on STO substrates and under various specific growth conditions (thickness t, pressure P_{O_2}): #1: 0% Co, $t = 150$ nm, $P_{O_2} = 10^{-6}$ mbar; #2: 1.5% Co, $t = 150$ nm, $P_{O_2} = 8 \cdot 10^{-7}$ mbar; #3: 1.5% Co, t = 150 nm, $P_{O_2} = 10^{-6}$ mbar; #4: 1.5% Co, $t =$ 20 nm, $P_{O_2} = 10^{-6}$ mbar. These experiments were done with an AC set-up (22 Hz), with magnetic fields up to $B = 16$ T and temperatures down to $T = 1.5$ K [6]. Our experimental data (cf. Fig. 2) show that the longitudinal resistance R_{xx} varies significantly with the temperature, and extremely large ratios of $R_{xx}(300\,\text{K})/R_{xx}(1.5\,\text{K}) \sim 700-1700$ were found. These samples are also characterized by large low-temperature electronic mobilities (up to 10^4 cm^2/V s, for $T < 10$ K). Consistently, such highly conductive systems exhibit Shubnikov–de Haas (SdH) oscillations in the field dependence of their magnetoresistance at high enough applied fields and low enough temperatures ($B \geqslant 6$ T, $T \leqslant 4$ K)—see Fig. 3. The period of these SdH oscillations is independent of the field direction (parallel or perpendicular to the film plane, as is shown in the inset of Fig. 3). This excludes any interface confinement of carriers and indicates that the high-mobility gas is three-dimensional.

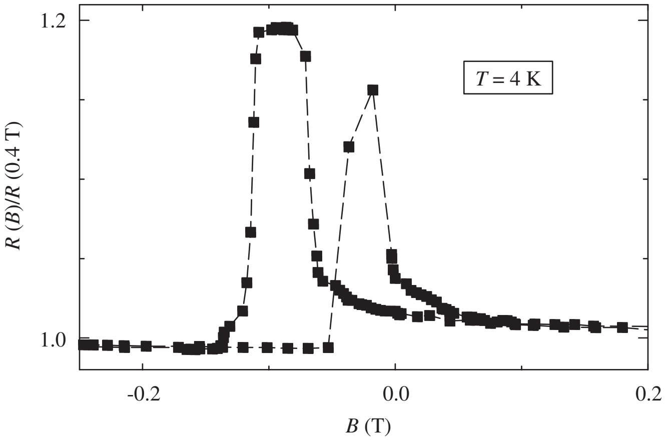

Fig. 1. Magnetic field dependence of the resistance of a Co-LSTO/LAO/Co magnetic tunnel junction at 4 K (the bias voltage was 50 mV).

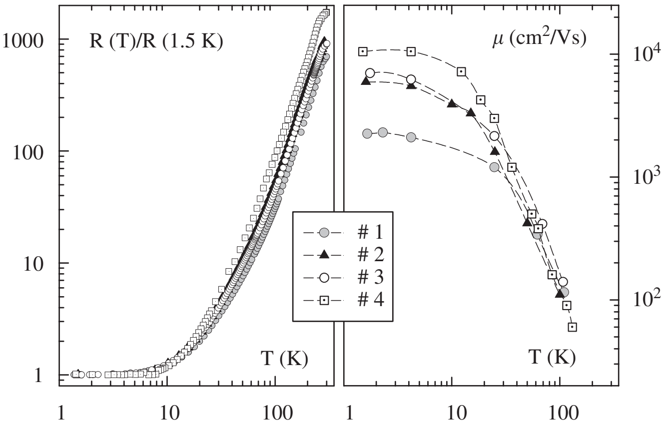

Fig. 2. Temperature dependence of the normalized resistivity (left) and of the mobility (right) for four different samples.

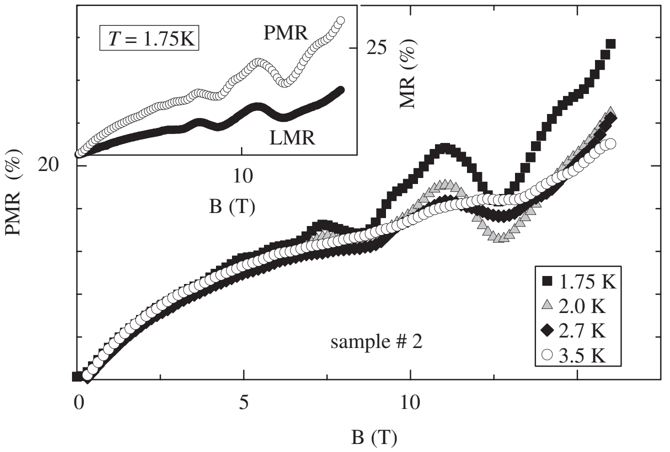

Fig. 3. Magnetic field dependence of the perpendicular magnetoresistance for sample #2. Inset: perpendicular (PMR) and longitudinal (LMR) magnetoresistance for the same sample at 1.75 K.

The thickness t_{hm} of the high mobility system was estimated combining the Hall effect and magnetoresistance experiments [6]. The Hall resistance R_{xy} at $B = 16$ T yielded the sheet carrier density $nt_{\mathrm{hm}} = (B/eR_{xy})$, where n is the carrier density, B the applied field, and e the electrical charge. On the other hand, the carrier density n could be determined *independently* through the analysis of the SdH oscillations, giving values of $n \approx 1.0 \times 10^{18}$–$1.9 \times 10^{18}\,\mathrm{cm}^{-3}$. Such an approach finally gave values of the thickness $t_{\mathrm{hm}} \approx 300$–$900\,\mu\mathrm{m}$.

High mobility and SdH oscillations in bulk STO single crystals (uniformly doped throughout their volume with a low amount of Nb-, La- or oxygen vacancies) are known for decades [7]. Thus, and in agreement with the above discussion, we suggest that the most likely origin of the high mobility in the Co-LSTO/STO samples is the doping of the STO substrate with oxygen vacancies (which is promoted by the low growth oxygen pressure and high temperature during deposition).

Finally, we should emphasize the great potential of the heterostructures combining high mobility STO with other oxides, because they could offer different functionalities for the realization of all-oxide electronic devices.

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