Growth and Transport Properties of [Rare Earth]TiO3/SrTiO3 Interfaces

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Citation

Lebedev, N. (2020, December 1). *Growth and Transport Properties of [Rare Earth]TiO3/SrTiO3 Interfaces*. *Casimir PhD Series*. Retrieved from https://hdl.handle.net/1887/138477

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Issue Date: 2020-12-01

Summary

Under particular circumstances, interfaces between two insulating oxide materials can become metallic. The ensuing two-dimensional electron system forms an intriguing object for research. This work focuses on the magnetotransport properties of conducting interfaces formed by a single crystalline, insulating and non-magnetic substrate, $SrTiO_3$ (STO), on which another insulating oxide with a similar crystal structure is deposited in the form of a thin film. This second material will in general contain magnetic ions. A special property of STO is that its large dielectric constant allows the substrate to be used as a back gate, allowing an electric field to vary and control the amount of charge carriers at the interface. The thin-film building blocks which were studied are similar to STO, also perovskites, namely the canonical material $LaAlO_3$(LAO) (insulating and non-magnetic) and the rare earth titanates $GdTiO_3$(GTO), $Eu_{1-x}La_xTiO_3$ (x=0 and 0.1), and $LaTiO_3$(LTO), all of which are magnetic. The main question I wanted to answer was whether the conduction electrons at the interface can become magnetic, that is spin-polarized, due to the use of the magnetic ions in the thin films. Moreover, several of the studied interfaces show superconductivity at low temperatures, which could also be investigated.

The thin film deposition was performed by Pulsed Laser Deposition (PLD). Not all materials are easy to grow in a reproducible manner in this way, but despite that, a wide range of phenomena such as the Anomalous Hall effect (AHE) (which is connected to the occurrence of magnetism), anomalous behavior of the magnetoresistance, non-uniform superconductivity, magnetic hysteresis, and signatures of the Kondo effect (yielding a minimum in the resistance when the temperature is decreased from room temperature) could be successfully studied. In the following, new insights deriving from this thesis are enumerated. Generically, all observations report that the conductance of the interface can be either due to one type (band) of carriers, or to two types or bands. The gate voltage tunes the Fermi energy and can bring the system from a one-band to a two-band regime, by injecting charge carriers and populating or depopulating the second band. The voltage at which the second band it switched on is called the Lifshitz point.

The Anomalous Hall Effect in magnetically doped oxide interfaces

Various perovskite oxide interfaces display the Anomalous Hall Effect. Its importance is that it is usually understood as a signature of the presence of ferromagnetism (long range magnetic order). Different mechanisms have been proposed to generate AHE, and there is a tendency to ascribe its observation to a one-ingredient mechanism despite the fact that, by its nature, it is the result of a complex interplay of different effects. To study the AHE, we chose delta-doped LAO/X/STO, with X being a few-atomic-layer thin magnetic oxide, either GTO or $Eu_{1-x}La_xTiO_3$(x=0, 0.1). It was experimentally difficult to grow GTO layers. LAO/GTO/STO was found to be severely intermixed and become $La_{1-x}Gd_xAlO_3/Gd_{1-y}La_yTi_{1-z}Al_zO_3$/STO (LGAO/GLTAO/STO), although still preserving crystallinity. As demonstrated in chapter 3, these samples exhibit an AHE which is tunable by the gate voltage and occurs at low temperatures in the two-band regime, close to the Lifshitz point. Two methods were utilized for the analysis, one which was proposed earlier in the literature, and one which was adapted from more conventional AHE analysis. They yielded similar results and indicated that the vicinity of the Lifshitz point and the ensuing strong spin-orbit coupling are important for observing the AHE. An important and unexpected result is that we did not find indications of ferromagnetism, which was probed directly via scanning SQUID microscopy. This implies that the system is rather superparamagnetic or paramagnetic rather than ferromagnetic. A follow-up study of AHE in LAO/$EuTiO_3$/STO (LAO/ETO/STO), LAO/$Eu_{0.9}La_{0.1}TiO_3$/STO (LAO/ELTO/STO), and LAO/STO heterostructures is presented in chapter 6. There, we did not observe AHE in the non-delta-doped sample, whereas the strength of AHE increases with increasing concentration of Eu. Although some authors argue that external magnetic doping need not be part of the mechanism of the AHE, here we see that magnetic doping is important for the AHE in LAO/ETO/STO structures. Intriguingly, a scaling analysis did not show the behavior expected for so-called skew-scattering, which is taken as the the main mechanism for AHE in most structures.

A percolative superconducting transition at the interface between an amorphous LTO film and a STO substrate

The interfaces that are discussed in this thesis are formed by perovkite titanates, deposited on STO. $LaTiO_3$is the compound which does not contain a magnetic Rare Earth ion, and therefore serves a reference system. The samples we grew show the LTO layer to be amorphous, not crystalline, which makes for an interesting interface system in itself, despite the somewhat uncontrolled nature of the amorphization process. This system exhibits the usual and typical properties of these oxide interfaces, namely two-band behavior and a Lifshitz point. In that sense, the amorphous system does not appear to be different from a crystalline one. However, a large linear magnetoresistance (MR) and a percolative superconducting transition indicate large inhomogeneities formed at the interface. To describe the superconducting transition, we used a resistor model, which describes the observations quite well. The (unevenly distributed) oxygen stoichiometry is argued to be a reason for the formation of large-scale inhomogeneities.

The coexistence of superconductivity and magnetism at oxide interfaces

The coexistence of superconductivity and magnetism at the conducting oxide interface is still a hotly debated topic. Initial reports on the signatures of magnetism in the superconducting state were met with excitement. Early studies on the magnetoresistance of such superconducting interfaces described hysteresis observed below the superconducting transition temperatures and discussed the possible role of magnetism. The non-trivial behavior, which could not be easily ascribed to ferromagnetic magnetization dynamics alone, was explained through a complicated picture of a superconductor in proximity to a complex magnetic texture. Other studies argued that the dependence of the MR effects on the sweep rate of the magnetic field indicate magnetocaloric effects rather than magnetic effects in the sample. Although such effects can indeed play a role, we find that magnetic hysteresis in the oxide interfaces can also be the sign of an inhomogeneous superconducting state. In chapter 5, two oxide systems with different types of disorder were studied. One system is the interface between amorphous LTO and crystalline STO, known from chapter 4; the other is the crystalline but strongly intermixed LGAO/GLTAO/STO system from chapter 3. The first one has disorder due to a nonuniform oxygen content, whereas the second has magnetic atoms that suppress superconductivity and create weak links between superconducting regions in the system. The amorphous system shows signatures of large-scale inhomogeneities, whereas a two-level model can describe LGAO/GLTAO/STO. Despite some differences, both systems exhibit magnetic hysteresis at low temperatures, which can be understood through the models for non-uniform superconductors without including ferromagnetism.

A Kondo-effect in the conductivity of oxide interfaces

In conducting oxide interfaces, a minimum in the temperature dependence of the sheet resistance, is often observed. For a long time it was considered as strong evidence of a Kondo effect, in which magnetic moments present at the interfaces were shielded by conduction electrons. Switching between a regime showing the AHE (and so ferromagnetism) and this regime was usually considered to be further evidence of the Kondo picture. Early pressure experiments proposed an alternative explanation which was overlooked in subsequent research. However, detailed experimental and theoretical studies of the temperature and gate dependence of the magnetoresistance showed the Kondo-picture to disagree with experiments. The current work also addresses this question and concentrates on the temperature dependence of the sheet resistance. As shown in chapter 6, a gate-tunable minimum in the temperature dependence of the sheet resistance was observed in all systems studied here. Through a detailed study of magnetotransport properties we show that the minimum turns out to be relatively insensitive to magnetic doping, which is not expected for the magnetic Kondo picture, whereas we do observe the apparent effects on the AHE that were mentioned earlier. As it turns out, other mechanisms such as the temperature and electric field dependence of the STO permittivity, charge trapping at the interface, the behavior of the carrier concentrations and mobilities of both conduction bands, and spinless scattering mechanisms are sufficient to describe the observed minimum. Even if the Kondo effect cannot be ruled out entirely, it most likely is masked by the effects mentioned above. This observation may shine new light on the sometimes confusing behavior of the magnetoelectric transport properties of oxide interfaces.