論 文 の 内 容 の 要 旨

論文題目　　Electronic Control of Transport Properties at Oxide Interfaces
（酸化物界面における輸送特性のキャリア制御）

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1. Introduction

Transition metal oxides are typical examples of strongly correlated materials. Many interesting physical properties of oxide materials are displayed from the layered nature of the crystal structure, exhibiting a broad range of functionalities related to chemical composition [1], superlattice [2], or interface design [3]. This characteristic can be enriched with artificially grown thin film heterostructures where different materials are combined either in single interfaces or in repetitive superlattices, allowing for systematic studies of two-dimensional oxide structures.

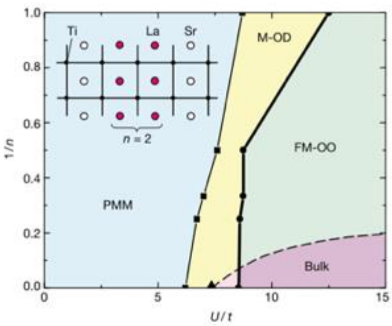

Fig.1 Spin and orbital phase diagram of $LaTiO_3$ / $SrTiO_3$ superlattices [4].

In this thesis work, I pay my attention to a delta-doped $LaTiO_3/SrTiO_3$ heterostructure and explore possible magnetic properties. Although $SrTiO_3$ itself is nominally a d^0 diamagnetic material, theoretical studies have suggested that various magnetically ordered phases may exist at the interface [4]. When $LaTiO_3$ layers of variable thickness are embedded in $SrTiO_3$, paramagnetic, metallic, and even ferromagnetic phases can appear in two-dimensional TiO_2 layers due to a preferential occupation of the d_{xy} orbitals (Fig. 1). Such spatially restricted orbital occupation can be achieved at heterostructure interfaces and in delta-doped quantum wells. This study plays an important role not only in the sense of fundamental interface physics but in terms of a strategy for application concerning magnetic materials without accompanying heavy elements.

Since possible magnetic states form at an interface embedded inside a crystal, common surface-sensitive analysis techniques cannot be used to probe for magnetism, while bulk measurements of magnetization are ineffective due to the small number of spins involved. I, therefore, attempt to probe the presence of magnetism by magnetotransport analysis while systematically varying the doping level of the quantum well either by changing the layer thickness or by electrostatic gating. I demonstrate here several ways of tuning the carrier density in this delta-doped system in a way that is easily applicable to spintronic device designs in typical transistor-like configurations.

The samples were grown on 0.2° miscut $SrTiO_3$ (001) single crystal substrates by pulsed laser deposition. An embedded layer of $LaTiO_3$ doped electrons into the $SrTiO_3$ substrate and $SrTiO_3$ capping layers were grown on top of the $LaTiO_3/SrTiO_3$ heterostructure in order to eliminate the effect of a surface depletion layer on the mobile carriers at the interface. The single-layer thickness control is achieved during film deposition with the help of feedback from reflection high-energy electron diffraction (RHEED) analysis. The substrates were pre-annealed at 900°C for ~30 minutes at an oxygen pressure of 10^{-6} Torr to obtain an atomically flat surface with a regular step-and-terrace morphology. The samples were heated with an infrared laser and kept at a deliberately low temperature of 500°C during the deposition to prevent La/Sr interdiffusion, leading to an asymmetric spread of carriers around the $LaTiO_3$ layer. Finally, the films were post-annealed in air at 400°C for 6 hours in a furnace to fill any oxygen vacancies created during the heterostructure growth. The surface morphology of crystals is measured by atomic force microscopy (AFM). If the final heterostructure surface is atomically flat, we can be sure that all the embedded interfaces are near-perfectly two-dimensional as well.

2. Transport and magnetotransport properties

The quantum wells are formed in the vicinity of the $LaTiO_3$ layer, and the thickness of the $LaTiO_3$ doping layer and the $SrTiO_3$ capping layer determine the number of carriers in the quantum well, the quantum well width, and the asymmetry of the confining potential. Since the thickness of a depletion layer is proportional to the square root of dielectric permittivity, carriers spread mainly into the single crystal substrate $SrTiO_3$, and any carriers that are doped into the low-crystallinity $SrTiO_3$ cap layer have sufficiently low mobility to be neglected in transport analysis.

All heterostructures with nominal $LaTiO_3$ layer thicknesses of over 0.5 unit cells(uc) were metallic with a fixed thickness of the $SrTiO_3$ cap layer at 10 nm. A transition to an insulating ground state occurs at an average $LaTiO_3$ coverage of about 0.3 uc, and adding $LaTiO_3$ layers induced more conductivity. A common feature of $SrTiO_3$ thin films is the phenomenon of surface depletion. Reducing the capping layer width will deplete carriers from the quantum well, leading to an insulating ground state. A gradual reduction of conductivity once the cap layer thickness is reduced below 20 uc. A semiconductor-like behavior can be observed for a 10 uc cap layer, but below this thickness, the heterostructures are highly insulating.

・Hall measurement

Hall effect measurements were used to analyze the carrier density and mobility at the interfaces. The measurements were done in a physical property measurement system (PPMS) using Hall bars cut into the heterostructures by mechanical milling. As expected [5], non-linear Hall resistance was observed at low temperature, which indicates that several types of carriers with different mobilities and densities were distributed along the *z* direction. The low-mobility carriers characterized by high *n* and low μ are confined to the interface, and another electron population, characterized by low *n* and high μ, spills out further into the single-crystal substrate from the interfacial quantum well.

・Magnetoresistance measurement

The magnetoresistance (MR) response was measured for different $LaTiO_3$ thickness samples. Metallic thin films in a perpendicular magnetic field are expected to monotonically show positive MR at low magnetic fields due to the cyclotron motion of carriers. On the other hand, no geometrical MR is expected when the magnetic field is applied parallel to the in-plane current flow. The appearance of strong MR in this geometry may thus be an indication of interfacial magnetic order.

The same Hall bar samples were used for MR measurements. In all samples, the MR increases with decreasing temperature while applying a magnetic field along the surface normal direction. Strong positive MR of up to 200% is seen in the thickest $LaTiO_3$ sample. The MR shape is parabolic only at low magnetic fields and becomes linear for higher carrier densities i.e., thicker $LaTiO_3$ layer. The slope of the MR curves at low field, which is proportional to carrier mobility, becomes sharper at lower temperatures or when adding $LaTiO_3$ layers. Considering the Hall analysis, this MR slope dependence can be interpreted as reflecting the increase of the carrier density of high-mobility carriers deep in the substrate.

When the magnetic field was applied parallel to the film, and the current flow direction, a large (~50%) negative MR was observed below 50 K. The MR increased rapidly in the low magnetic field region and saturated at high fields. The low-field slope is proportional to the $LaTiO_3$ film thickness (i.e., carrier density) and can be related to the filling of the preferred d_{xy} orbitals at the interface [6]. Theoretical and experimental studies [6,7] have suggested that there is a depth profile of orbital occupancy near the interface with carriers preferring the Ti $3d_{xy}$ states at the interface while uniform $3d_{xz,yz}$ occupancy occurs in the $SrTiO_3$ bulk. This orbital order of the different subbands is considered as the origin of spin-orbit interaction in this system.

3. Back-gating experiment

The electric field effect can be used for fine-tuning of the carrier density or the potential profile of a semiconductor quantum well. In a typical field-effect transistor structure, the electric field is applied to an electrode formed on top of the heterostructure. However, since $SrTiO_3$ has a dielectric permittivity of over 2×10^4 below 10 K, it is possible to use back gating by applying a bias on an electrode formed on the back of a 0.5 mm thick substrate. This method avoids gate leak current problems and is more suitable for quantum well shape tuning.

Experimental results show that a negative back gate voltage on the interface (which is held close to 0 V) can compress the quantum well, resulting in an increase of sample resistance by up to three times at $V_g = -100$ V at 4 K due to disorder near the interface. This behavior is reversible, which implies that the total number of electrons is constant.

A positive gate bias is expected to broaden the quantum well and may remove electrons from the well. A broader well allows current to be carried in the deeper defect-free region in the substrate, reducing the sample resistance. However, the confining potential barrier may reach the Fermi level, at which point the resistance change saturates. Moreover, strong positive bias has been observed to allow electrons to escape from the well and be irreversibly trapped, presumably in the substrate. As a result, once high positive bias is applied, the heterostructure resistance increases irreversibly, and a memory effect appears, implying that there are still remaining impurities even after the long post-annealing process. The detrapped carriers can be reset by warming up a sample to around 130 K or by illuminating with 300~1300 nm light.

Trapping carriers affect in-plane magnetotransport behavior. The intensity of in-plane negative MR is suppressed by the order of ten, which may indicate that not only the interface carriers but deep carriers are also responsible for the interfacial magnetic ordering.

4. Conclusions

In this study, delta-doped $LaTiO_3/SrTiO_3$ heterostructures were fabricated, characterized, and controlled. With the samples varied with $LaTiO_3$ and $SrTiO_3$ thickness, magnetoresistance and Hall effect measurements were performed at 2 ~ 300 K in different applied magnetic field configurations. Non-linear Hall resistance, positive out-of-plane magnetoresistance, and strong negative in-plane magnetoresistance were observed below 50 K. Non-linearity of Hall resistance was analyzed with the help of a two-carrier model that applies to systems with several types of carriers that contribute to the current flow in a parallel circuit configuration. The magnetoresistance response was investigated in different magnetic fields. The positive out-of-plane magnetoresistance was parabolic at low magnetic fields and became linear for high fields. In contrast, the strong negative in-plane magnetoresistance observed, possibly due to the interface spin-orbit interaction. The electric field effect was used to tune the carrier confinement potential profile of the heterostructure. By applying a negative bias voltage, the quantum well can be compressed, and the sample resistance grows rapidly. A positive gate bias may broaden the quantum well, which induced the spilling of carriers out of the interfacial quantum well and trapping of carriers deeper in the substrate, leading to only a decrease of resistance. Electrons that escaped from the quantum well could not be recovered by negative biasing, and a strong irreversible resistance change was observed. All measurement results can be enhanced or suppressed by changing doping layer thickness or by applying gate bias.

References

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