研究成果の概要:

We have developed a process for fabricating micron-scale fully epitaxial top-gate oxide field-effect transistors that use an oxide channel, oxide source and drain electrodes and a wide-gap oxide gate insulator. We have studied charge accumulation at CaHfO$_3$/SrTiO$_3$, DyScO$_3$/SrTiO$_3$, and SrTiO$_3$/LaTiO$_3$ interfaces. As a way to confine carriers in a narrower layer at an interface, we are developing Ruddlesden-Popper-type two-dimensional quantum wells.

<table>
<thead>
<tr>
<th>年度</th>
<th>直接経費</th>
<th>間接経費</th>
<th>合 計</th>
</tr>
</thead>
<tbody>
<tr>
<td>2007年度</td>
<td>[経費]</td>
<td>[経費]</td>
<td>[総計]</td>
</tr>
<tr>
<td>2008年度</td>
<td>[経費]</td>
<td>[経費]</td>
<td>[総計]</td>
</tr>
<tr>
<td>年度</td>
<td>[経費]</td>
<td>[経費]</td>
<td>[総計]</td>
</tr>
<tr>
<td>年度</td>
<td>[経費]</td>
<td>[経費]</td>
<td>[総計]</td>
</tr>
<tr>
<td>総 計</td>
<td>[経費]</td>
<td>[経費]</td>
<td>[総計]</td>
</tr>
</tbody>
</table>

研究分野：数物系科学
科研費の分類・細目：物理学・物性
キーワード：電子デバイス・機器、ナノ材料、表面・界面物性、物性実験

1. 研究開始当初の背景
Many oxides have very rich electronic phase diagrams with various insulating, metallic, magnetic, superconducting, etc. phases. In many materials, these phase transitions can be controlled by changing the density of charge carriers, i.e. doping. A particularly attractive method of doping oxide is to use the electrostatic field effect to tune the carrier density in a

two-dimensional layer continuously over a phase transition.

2. 研究の目的
The purpose of the project was to study the possibility of observing phase transitions in SrTiO$_3$ and other oxides by constructing fully epitaxial field effect transistors, in which the carrier density in the transistor channel can be adjusted by simply
changing the gate voltage. Since the carrier density modulation range is limited by the breakdown field of the insulator, our purpose was to study phase transitions that occur at moderately low carrier densities, such as the insulator-metal and insulator-superconductor transition in SrTiO$_3$. Of particular interest for us was the possibility of controlling localization-driven insulator-metal transitions in two-dimensional oxide layers.

3. 研究の方法
We used laser molecular beam epitaxy to grow high-quality fully epitaxial oxide heterostructures consisting of SrTiO$_3$ and several different insulator materials (CaHfO$_3$ and DyScO$_3$) and different channel compositions (SrTiO$_3$, oxygen-deficient SrTiO$_3$, (La,Sr)TiO$_3$, and various Ruddlesden-Popper type manganites. The heterostructures were patterned by photolithography and ion milling. Transistor electrodes consisting of metallic oxygen-deficient SrTiO$_3$ were also made by ion milling. The transport properties of the field-effect transistors and magnetotransport of heterostructures were measured as a function of temperature, carrier density, magnetic field, and ultraviolet illumination. The occurrence of metal-insulator transitions was observed.

4. 研究成果
(1) We developed a process for fabricating epitaxial field-effect transistors based on epitaxial oxide layers (Fig. 1). This device structure can be used to study the transport properties of various ultrathin oxide layers that can be grown on SrTiO$_3$. The best performance is generally obtained in the smallest devices, which are less affected by defects in the films.

![Fig. 1 Array of transistors with various channel sizes.](image1)

(2) A technique was developed to use oxygen-deficient SrTiO$_3$ as an electrode material for epitaxial oxide FETs (Fig. 2). A key feature of this design is that the channel interface layer can be grown before any other device processing or patterning. In this way, the best channel interface quality can be obtained.

![Fig. 2 Cross-sectional view and optical image of an oxide transistor.](image2)

The SrTiO$_3$-based oxide FETs were used in depletion mode for mapping the density of in-gap states at the SrTiO$_3$/DyScO$_3$ interface. The variation of the activation energy of channel conductance as a function of gate bias, or quasi-Fermi level distance from the conduction band mobility edge, shows plateau-like behavior about 400 meV below the conduction band bottom (Fig. 3). This result was obtained in a slightly oxygen-deficient crystal, showing that simple transport measurements in an FET can provide electronic density information that would normally require synchrotron source photoelectron emission spectroscopy measurements. Additionally, the FET measurements probe the electronic density in an essentially two-dimensional interface layer, which is generally not accessible in photoemission experiments.

![Fig. 3 Schematic diagram of in-gap states near SrTiO3 channel band and measured density of states.](image3)

The ability to induce a large enough carrier density at an interface to observe phase transitions in common oxide systems, is limited by the breakdown of the insulator and trapping of charge at interfaces. Charge trapping is also the basis for some types of resistive switching devices that have been proposed as new high-density non-volatile memories.
Charge trapping at various interfaces in an oxide device can be studied by capacitance-voltage analysis (insulator layers) and by measuring the temperature dependence of the transistor switching characteristics (channel material). Since the mobility of charge carriers in SrTiO$_3$ is a strong function of temperature, measuring the low-temperature performance of an FET can clearly show if interface traps (and thus defects) remain at the channel interface. An example of such a measurement is shown in Fig. 4.

![Fig. 4 Temperature dependence of SrTiO$_3$ / DyScO$_3$ FET switching characteristics.](image)

The data in Fig. 4 shows that the off-current of the device drops exponentially with temperature. This is caused by in-gap defect states in SrTiO$_3$, assigned mostly to oxygen defects. The increase of the slope of the switching curves at low temperature shows that the mobility increases at low temperature, as expected for SrTiO$_3$.

The mobility behavior of a FET is a good probe of interface quality in an electronic sense. The field-effect mobility measured in a 10 μm channel length device is overlaid on bulk mobility data for SrTiO$_3$ in Fig. 5. This type of measurement sees the average mobility in the channel region and is generally lower than the bulk mobility. For the particular device for which the data is shown in Fig. 5, it is clear that the mobility does follow the bulk characteristics and it can be concluded that epitaxial oxide interfaces can be grown with sufficiently low impurity or structural defect densities. This is important if we consider an FET as a general tool for measuring electronic phase diagrams in nm-scale oxide layers.

![Fig. 6 Illustration of a metal insulator transition in SrTiO$_3$.](image)

We have observed metal-superconductor and metal-insulator transitions in oxide FETs and related structures. The main benefit of an FET as an electronic doping tool is that the electron density can be changed reversibly, and with much better accuracy than is possible by chemical doping. It is therefore possible to study the effects of small perturbations caused by changes in temperature, magnetic fields, light, etc. on a material at the ‘midpoint’ of a phase transition. A metal-insulator transition measured in a SrTiO$_3$ FET is illustrated in Fig. 6.

The data in Fig. 6 shows how the sheet resistance behavior vs. temperature changes as the electric field is varied between 0.8 and 1.3 MV/cm. The maximum field in this case corresponds to a sheet carrier density increase of about 5x10$^{12}$ cm$^{-2}$. Considering that for bulk SrTiO$_3$, the transition occurs at a carrier density of about 5x10$^{17}$ cm$^{-3}$, it is evident that the conducting channel thickness is on the order of at least tens of nm.

The thickness of a conducting layer at an oxide heterointerface is determined by the depth distribution of defects and the dielectric properties of the material. In the materials studied in this project, it is important to consider the motion of oxygen vacancies, which can have very high mobilities at typical thin film growth temperatures of about 700 °C.
In order to confine carriers in a thinner interface layer and thus achieve higher volume densities of carriers, it is necessary to restrict the movement of either cation or anion defects and electrons. One possible approach is to use rocksalt-type layers to isolate perovskite layers from the rest of the crystal, i.e. a structure similar to a unit cell of a Ruddlesden-Popper phase.

Fig. 7 shows the specular intensity oscillations during laser molecular beam epitaxy growth of a three unit cell thick film of (La,Sr)₇(Mn,Ru)₃O₁₅. The data shows that it is possible to determine exactly which crystal blocks grow on the surface and the layer sequence can be changed by growing a film a single block at a time, using separate A and B-site cation targets.

(8) In order to achieve tighter electron confinement, we have studied this in SrTiO₃ / LaTiO₃ / SrTiO₃ trilayer structures, where the LaTiO₃ layer coverage is less than 1 full unit cell. This type of a fractional-layer quantum well shows strong localization and a transition from a metallic or semiconducting state to an insulating state at about 100 K. The temperature dependence of resistivity of fractional-layer LaTiO₃ quantum wells is shown in Fig. 8. LaTiO₃ layers that are close to 1 unit cell thick, are metallic. At a coverage of about 0.2 unit cells, a sharp transition to insulating state is seen at about 50 K. This type of well structures can also be used for doping studies in FET structures, since the quantum wells can easily be integrated in the FET channel.

5. 主な発表論文等
(雑誌論文)(計 14件)
4. 大西 剛, パルスレーザー堆積法による複酸化物薄膜のエピタキシー, 機能性材料 28 (2008) 6-14, 論文誌有。

(学会発表) (計 29件)
1. 大西 剛, Mikko Lippmaa, SrTiO₃の表面・界面電子伝導: 2 層構造モデル, 第 56 回応用物理学関係連合講演会 2009.3.30-4.2 (筑波大)
2. 伊高健治, Lippmaa Mikko, 大西 慶, 片山正士, 知京秀裕, 鶴沼秀臣, フレキシブル真空発生機構を備えた超小型モジュール型薄膜・評価測定システムの開発, 第 56 回応用物理学関係連合講演会 2009.3.30-4.2 (筑波大)
3. 大塚奈奈, 西尾和記, ミッコ マトヴィエフ, ミック リップマー, SrTiO₃にはさまれた複層 LaTiO₃ 膜における輸送特性, 第 56 回応用物理学関係連合講演会 2009.3.30-4.2 (筑波大)
4. 菊月連也, 大西 剛, ミック リップマー, VO2 薄膜における歪みを利用した相転移現象, 第 56 回応用物理学関係連合講演会 2009.3.30-4.2 (筑波大)
5. 西尾和記, 安部拓也, ミック リップマー, DyScO₃/SrTiO₃表面の電気導電特性, 第 56 回応用物理学関係連合講演会 2009.3.30-4.2 (筑波大)
6. M. Matvejeff, T. Chikyow, M. Lippmaa, "Growth of (ultra) thin La₂Sr₁₂Mn₂O₁₉ + Ru₂O₅/RuO₂ thin films on SrTiO₃", 15th Workshop on Oxide Electronics, 14-17.9.2008, Estes Park, USA
8. 小塚裕介, 大西 剛, Mikko Lippmaa, 定 Riffee, Harold Hwang, バランスレーザー堆積法による低濃度キャリアドープされた高移動度 n 型 SrTiO₃薄膜の製作, 第 69 回応用物理学学会学術講演会 2009.9.2-5 (中部大)
9. 安部拓也, 西尾和記, 大西 剛, ミック リップマー, 界面層が DyScO₃ キャリアの誘電特性に及ぼす影響, 第 69 回応用物理学学会学術講演会 2009.9.2-5 (中部大)
10. 西尾和記, 安部拓也, 大西 剛, ミック リップマー, 単結晶 SrTiO₃ 電界効果トラシスタにおける電流の緩和現象, 第 69 回応用物理学学会学術講演会 2009.9.2-5 (中部大)
11. ミッコ マトヴィエフ, ミック リップマー, 単結晶 SrTiO₃ 上への La₂Sr₁₂Mn₂O₁₉十 Ru₂O₅十ポッパー極薄膜成長, 第 69 回応用物理学学会学術講演会 2009.9.2-5 (中部大)
12. 西尾和記, 安部拓也, 佐藤泰輔, 魚住篤之, 津矢圭介, 大西 剛, リップマーミッカ, ディプローショング型電界効果トラシスタによる SrTiO₃パンド間距離の評価, 第 55 回応用物理学関係連合講演会 2008.3.27-30 日本大学
13. 梅野 Antarctic, 根田尚之, 太久保勇男, 接頭しての広川, 大西 剛, 杉本任司 Mikko Lippmaa, 鶴沼秀臣, 尾崎正治, 人工界面酸化物層を導入した金属/Pr₀.₇Ca₀.₃MnO₃ 界面の電流-電圧特性, 第 55 回応用物理学関係連合講演会 2008.3.27-30 日本大学
15. 大西 剛, 望月圭介, 山本博文, 藤本英司, 角谷正友, Mikko Lippmaa, SrTiO₃ の
発表，第55回応用物理学関係連合講演会，2008.3.27-30，日本大学
23. 西尾和記，安部拓也，大西剛，山本剛久，Mikk Lippmaa, 单結晶SrTiO$_3$電界効果トランジスタに向けたエピタキシャルDyScO$_3$薄膜の作製，第68回応用物理学会学術講演会，2007.9.4-8，北海道工業大学
24. 大西剛，望月圭介，米澤卓三，上膳明良，Mikk Lippmaa, SrTiO$_3$の色，第68回応用物理学会学術講演会，2007.9.4-8，北海道工業大学
25. 大西剛，Mikk Lippmaa, Pulsed Laser Deposition：酸化物ターゲット中の酸素第68回応用物理学会学術講演会，2007.9.4-8，北海道工業大学
26. 浦田康文，大西剛，山本博文，石井誉，川南修一，Mikk Lippmaa，格子欠陥のあるSrTiO$_3$の光励起電気伝導特性，第68回応用物理学会学術講演会，2007.9.4-8，北海道工業大学
27. Mikk Lippmaa, 大西剛，西尾和記，安部拓也，単結晶SrTiO$_3$電界効果トランジスタに向けたDyScO$_3$薄膜の低温誘電特性，第68回応用物理学会学術講演会，2007.9.4-8，北海道工業大学

（図書）（計1件）