研究課題別評価書

1．研究課題名

静電エネルギーの発散を利用した人工界面相の創成と制御（Creation and Control of Artificial Interface Phases Induced by Tunable Electrostatic Divergences）

2．氏名

Harold Y. Hwang

3．研究のねらい

A central goal of modern materials research and nanoscience is the control of materials and their interfaces to atomic dimensions. In the search for ever higher functionality in solid state devices, transition metal oxides are fascinating systems which host a vast array of phenomena, such as superconductivity, magnetism, and ferroelectricity. To progress towards integrated devices using these materials, a fundamental understanding of interface electronic and atomic structure, barrier formation, and control must be developed. Our research centers on atomic scale oxide heterostructures (Fig. 1), with the aim of creating novel states and physical properties unobtainable in bulk materials. By atomic engineering the electrostatic boundary conditions at interfaces, we create novel functional interface states, opening a new frontier in interface science and device technology.

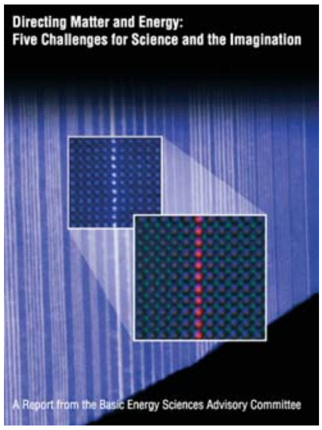

Fig. 1. Cover of the U.S. Dept. of Energy report "Directing Matter and Energy: Five Challenges for Science and the Imagination", based on our research.

4．研究成果

Tunable (super-)conducting interfaces at polar discontinuities. This proposal was based on our unexpected finding of a conducting interface between two band insulators, $LaAlO_3$ and $SrTiO_3$ [*Nature* **427**, 423 (2004)]. This conducting electron gas only occurred for one specific atomic termination between these two materials, which is a result of the sign of the polar discontinuity formed between them [*Nature Materials* **5**, 204 (2006)]. Based on these results, we have further explored the generality of this principle in other systems, in particular the $LaVO_3/SrTiO_3$ Mott insulator/band insulator heterointerface for various atomic configurations. The (001)-oriented n-type $VO_2/$*LaO*$/TiO_2$ polar discontinuity is conducting, exhibiting a $LaVO_3$ thickness-dependent metal-insulator transition and low temperature anomalous Hall effect. The (001) p-type $VO_2/$*SrO*$/TiO_2$ interface, formed by inserting a single layer of bulk metallic $SrVO_3$ or SrO, drives the interface insulating. The (110) heterointerface is also insulating, indicating interface conduction arising from electronic reconstructions.

The $LaAlO_3/SrTiO_3$ interface was remarkably found to be superconducting by the Geneva group [N. Reyren *et al.*, *Science* **317**, 1196 (2007)]. Using a back-gate geometry, we found that we could form a superconducting transistor which suppressed superconductivity at both positive and negative gate bias. Specifically, a 2D transition to an insulating state was found in depletion mode, and a transition to a metallic state in accumulation mode. Using magnetotransport studies of a gated structure, we find that the mobility variation is almost 5 times that of the sheet carrier density. Gate depletion strongly reduced the carrier mobility, as the electrons are pressed against the disorder of the interface barrier, dictating that 2D superconductivity was confined to the dirty limit. Motivated to overcome this, we have developed $SrTiO_3$ delta-doped heterostructures which exhibit 2D superconductivity (Fig. 2) with sufficiently high mobility to show 2D quantum oscillations in the normal state, giving the first experimental access to artificial 2D superconductors in the clean limit. These results suggest that delta-doped $SrTiO_3$ provides a model system to explore the quantum transport of both superconducting and normal electrons, and their interplay.

Modulation hole doping. All of the examples described above only create electrons at

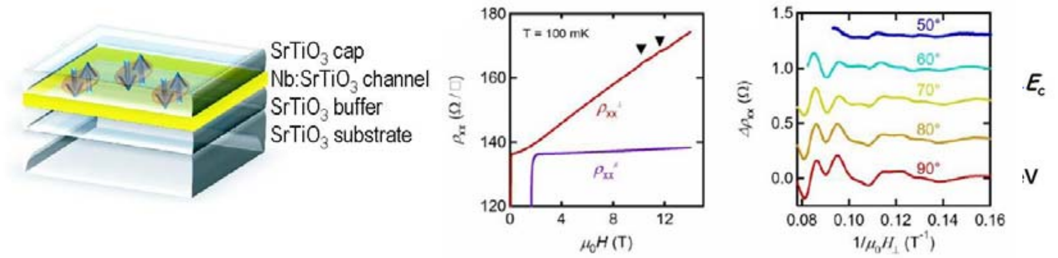

Fig. 2. Left: A sketch of the delta-doped heterostructure with a 5.5 nm superconducting channel. Center: Magnetic field dependence of the resistance of the device in perpendicular and parallel orientations, showing first a transition out of the superconducting state, and at high fields (arrows), quantum oscillations. Right: Scaling of the quantum oscillations with the perpendicular magnetic field, demonstrating two-dimensional one-electron states.

Fig. 3. Schematic quantum well of the correlated insulator $LaVO_3$ embedded in $LaAlO_3$, where the polar surface of the $LaAlO_3$ dopes the well when brought within a few nanometers.

interfaces. There has been a long-standing search for a method to similarly create a hole gas at interfaces. Figure 3 shows our realization of the first experimental example of polar hole doping in a quantum well using the Mott insulator $LaVO_3$ embedded in $LaAlO_3$, a heterostructure that is initially completely insulating. Although there are no internal polar discontinuities at the interfaces, when the polar surface is brought in close proximity to the active region, we can "modulation dope" holes in the quantum well, analogous to offset doping in GaAs high electron mobility transistors. This effect has been demonstrated in a series of experiments using transport, thermopower, and photoemission spectroscopy. This structure spatially separates the source of doping from the electrically active interface, providing a way to greatly reduce interface scattering and disorder. Furthermore, these results suggest a general method for tunable hole doping in oxide heterostructures in systems with accessible higher oxidation states.

Engineering Schottky junctions. The results described above lead naturally to the question whether we can incorporate these ideas of atomic engineering interfaces in devices. The model system we have used to develop devices is the Schottky diode. Using a variety of techniques, including current-voltage, capacitance, and internal photoemission spectroscopy, we have studied fundamental aspects of oxide Schottky barriers. When using $SrTiO_3$ as the doped semiconductor, a key feature is the strong temperature dependence of the barrier due to the changes in the permittivity, leading to novel phenomena such as polarity reversal in Au/Nb:$SrTiO_3$ junctions. By placing discrete impurities at the interface between $SrRuO_3$ and Nb:$SrTiO_3$, a resonant state can be induced, creating a regime of negative differential resistance. By changing the termination of the interface between (La,Sr)MnO_3 and Nb:$SrTiO_3$, the barrier height can be engineered to vary 0.5 eV between the same materials.

These and other developments in our understanding and control of oxide Schottky junctions gives us the basic tools needed to create more sophisticated devices. As a first step in this direction, we have used an all-perovskite heterostructure to create a hot electron transistor using back-to-back Schottky junctions, with a ferromagnetic metallic manganite as the base (Fig. 4). A key feature to successfully realize this device was the atomic engineering of the two active interfaces, which enabled the control of reverse bias leakage. A next goal is to incorporate magnetic field control of transistor function, which should be enabled by engineering magnetically active Schottky barrier heights, as we have previously shown for single interface devices. These results suggest that this geometry provides a fundamentally new approach to multifunctional 3-terminal devices in oxide heterostructures.

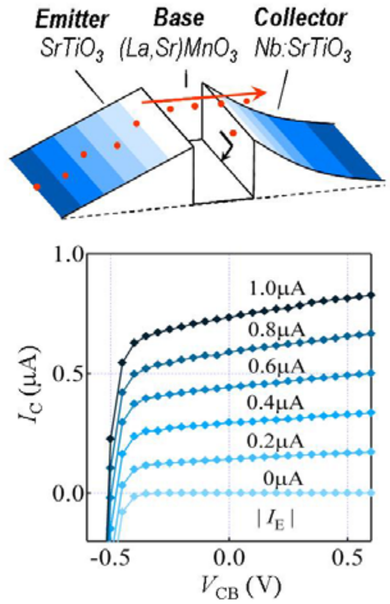

Fig. 4. Top: Schematic oxide heterostructure hot electron transistor. Bottom: Common base output characteristics at room temperature.

5. 自己評価

We have studied perovskite oxide thin films, interfaces, and multilayers with the general aim of developing complex oxide heteroepitaxial structures with new physical properties and device potential. Overall, we have made significant progress well aligned with the research aims of this proposal. The primary new heterostructure systems developed were vanadates and delta-doped $SrTiO_3$. Undoped manganites such as $LaMnO_3$, however, were not fully successful thus far due to a much more complex defect chemistry, which we continue to investigate. The electronic and spectroscopic characterization of these heterostructures has been quite successful, in part due to a growing network of collaborations, in addition to rapid development in our laboratory. In the aim to move closer to device concepts and engineering, we have made significant progress in the understanding of the electronic structure of Schottky junctions, as well as the ability to control their properties by atomic scale design. In summary, this should be a promising new direction for manipulating the many bulk physical properties associated with heterovalent transition metal oxides, applying the concept of bandgap engineering in perovskite heterostructures. In addition to a number of scientific accomplishments within this project, we believe this research has seeded several exciting new research concepts and directions, such as hot-electron transistors and delta-doping in oxides, which we will continue to pursue in future research.

6. 研究総括の見解

Oxide engineering to control the band structure has developed significantly in these few years, where Prof. Hwang is the one of the few players leading this world of science. It is quite impressive to see the invention of new materials systems such as vanadates very new to the oxide thin film work. An important result towards 3 terminal devices was performed by the success to demonstrate the Schottky junctions. As device materials are usually seriously concerned, people gradually may start to analyze the details of the electronic and geometric structures at the 2D interfaces.

7. 主な論文等

A. さきがけ個人研究者主導で得られた成果で主なもの

①論文(3 報)

1. M. Takizawa, Y. Hotta, T. Susaki, Y. Ishida, H. Wadati, Y. Takata, K. Horiba, M. Matsunami, S. Shin, M. Yabashi, K. Tamasaku, N. Nishio, T. Ishikawa, A. Fujimori, and H. Y. Hwang, "Spectroscopic Evidence for Competing Reconstructions in Polar Multilayers $LaAlO_3/LaVO_3/LaAlO_3$," *Physical Review Letters* **102**, 236401:1-4 (2009).
2. Y. Kozuka, M. Kim, C. Bell, B. G. Kim, Y. Hikita, and H. Y. Hwang, "Two-Dimensional Normal-State Quantum Oscillations in a Superconducting Heterostructure," *Nature* **462**, 487-490 (2009).
3. C. Bell, S. Harashima, Y. Kozuka, M. Kim, B. G. Kim, Y. Hikita, and H. Y. Hwang, "Dominant Mobility Modulation by the Electric Field Effect at the $LaAlO_3/SrTiO_3$ Interface," *Physical Review Letters* **103**, 226802:1-4 (2009).

②受賞(1 件)

1. 2008 IBM Japan Science Prize (Physics).

③招待講演(2 件)

1. "Modulation Doping of Electrons and Holes at Oxide Interfaces," American Physical Society March Meeting, Pittsburgh, PA, March 16-20, 2009.
2. "Interface between Magnetic and Nonmagnetic Oxides," Materials Research Society Spring Meeting, San Francisco, CA, April 13-17, 2009.

B. 本研究課題に関連した成果で主なもの

①論文(2報)

1. N. Ogawa, K. Miyano, M. Hosoda, T. Higuchi, C. Bell, Y. Hikita, and H. Y. Hwang, "Enhanced Lattice Polarization in $SrTiO_3/LaAlO_3$ Superlattices Measured Using Optical Second Harmonic Generation," *Physical Review B (Rapid Communications)* **80**, 081106:1-4 (2009). Editors' Suggestion.
2. Y. Kozuka, M. Kim, C. Bell, B. G. Kim, Y. Hikita, and H. Y. Hwang, "Two-Dimensional Normal-State Quantum Oscillations in a Superconducting Heterostructure," *Nature* **462**, 487-490 (2009).

②受賞(1 件)

1. 2008 IBM Japan Science Prize (Physics).

③招待講演(1件)

1. "Optical Control of the Electron Dimensionality and Localization in Photocarrier-Doped $SrTiO_3$, $KTaO_3$," Symposium for Emergent Materials Research, Pohang, Korea, May 23-24, 2008.