

An Ultra-Pure Heterostructure of a Transition metal Oxide.

Observation of the Quantum Hall Effect in an Oxide Heterostructure

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Recommended with a Commentary by A. P. Ramirez, *Bell Labs*.

With the end to CMOS scaling in sight, and the continued need for multifunctional sensors and sources, there is much research on devices whose functionality derives from nanoscale processes. The materials for such devices lie beyond traditional covalently-bonded semiconductors. They include such exotic structures as nanotube assemblies, molecular crystals and polymers, and lamellar heterostructures of nontraditional combinations of materials. Among the most exciting candidate materials for novel devices are the transition metal oxides. Oxides demonstrate most of the important collective behavior in crystalline systems, in particular behavior that elicits large (giant, and even colossal) responses to external fields. The prospect of realizing novel device functionality through exploiting such collective behavior is appealing and recent work by a group at Tohoku University has achieved a milestone along this path with the observation of the quantum Hall effect (QHE) in ZnO/Mg<sub>x</sub>Zn<sub>1-x</sub>O heterostructures.

Zinc oxide is, in bulk form, an important material with commercial applications in piezoelectric transducers and transparent thin films. It has a direct band gap of 3.3 eV and an exciton binding energy of ~60 meV, making it a candidate for short wavelength LEDs and lasers. High-quality crystals of ZnO, which is typically *n*-type in bulk, became available about 10 years ago. This enabled the identification of the point defects responsible for electron doping, and several groups have demonstrated *p*-type doping of ZnO. Thus, in addition to having useful bulk properties, ZnO is a good candidate in which to search for new device functionality. Recently the group at Tohoku University used nitrogen as the *p*-type dopant to grow, by MBE, *p-i-n* junctions that produced violet electroluminescence, a key step towards electrically pumped lasing. ZnO has also attracted a great deal of attention since the prediction of room-temperature ferromagnetism upon 5% Mn substitution for Zn in the wurtzite structure<sup>1</sup>. Despite experimental evidence for a ferromagnetic state, both theory and experiment on this subject are controversial at present<sup>(2,3)</sup>.

The ability to take a bulk semiconductor and create high performance devices such as transistors, photocells, and photodiodes, relies on the ability to control atoms and their resulting polarity discontinuities at interfaces. The multivalent ionic character of transition metals in oxides presents an additional device design parameter. As noted by H. Hwang<sup>4</sup>, bulk transition metal oxides can be thought of as nanoscopic heterostructures of various dimensions and polarity discontinuities, leading to charge transfer and a type of intrinsic modulation doping. In a kinetically-grown heterostructure, one has the ability to engineer the polarity discontinuity at the interface and achieve valence-state reconstruction. In such a device, it might even be possible to achieve

artificial Verwey phases from a single ionic type by appropriately tuning the layer structure<sup>5</sup>. Multivalency can also be a source of disorder, but recent work on LaAlO<sub>3</sub>/SrTiO<sub>3</sub> heterostructures showed it possible to keep disorder at the interface low enough to support quantum oscillations in the electron gas formed at the interface.

The present work builds on years of sustained research, both at Tohoku and elsewhere, aimed at fabricating high mobility epitaxial thin films of oxide materials and ZnO in particular. An important recent advance in ZnO films by the Tohoku group was the attainment of mobility exceeding that in the purest single crystals. The recipe for achieving low defect density in ZnO is as follows: 1) start with a single crystal substrate of ScAlMgO<sub>4</sub>, which is well lattice-matched to ZnO; 2) grow by laser ablation an atomically-flat and strain-free buffer layer of Mg<sub>x</sub>Zn<sub>1-x</sub>O at high-temperatures obtained by laser-heating the substrate; 3) grow the ZnO using a high-purity single crystal as the ablation target. A key to this process is laser-heating of the substrate, a technique also developed at Tohoku, which enables growth in arbitrary oxygen atmosphere without the oxidation problems of usual heating methods.

The polarity discontinuity achieved at the interface between ZnO and Mg<sub>x</sub>Zn<sub>1-x</sub>O is calculated by the authors to produce a finite free carrier density at  $x \sim 0.1$ . In the samples studied,  $x = 0.15 - 0.20$ , and the carrier densities ( $0.66 - 3.7 \times 10^{12} \text{ cm}^{-2}$ ) are measured to be in good agreement with the calculations. Electron mobilities of  $2,700 - 5,500 \text{ cm}^2/\text{Vs}$  at 1K were found and measurements of  $\rho_{xx}$  and  $\rho_{xy}$  as a function of magnetic field at 45 mK show clearly-defined QHE signatures, down to  $\nu = 2$  in one sample. Comparative measurements on the three samples used in this work raise a number of questions about spin degeneracy lifting by interface asymmetry. Having feedback from such an exquisitely sensitive probe of disorder as the QHE will help in understanding the nature of defects at the interface. In addition, zinc is a common non-magnetic substituting atom in magnetic materials so it's germane to ask whether in Zn itself can be substituted by any of the *3d* transition metals.

In the Tohoku experiment, the promise of oxide devices was not realized per se. The device performance doesn't employ a strongly correlated bulk state, and the physical effects are not new, having been observed since the early 80's in silicon MOSFETS. The importance of the experiment lies in its demonstration that a particularly delicate many-body state only found in the highest quality heterostructures, can also be realized in an oxide system – thus oxide interfaces can be made extremely pure. Knowing that high mobility devices can be made from one oxide compound semiconductor will provide greater justification for pursuing similar high device quality in other compound systems for both science and technology.

## References

- <sup>1</sup> T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, *Science* **287**, 1019 (2000).
- <sup>2</sup> D. Iusan, B. Sanyal, and O. Eriksson, *Physical Review B* **74** (2006).
- <sup>3</sup> J. M. D. Coey, M. Venkatesan, and C. B. Fitzgerald, *Nature Materials* **4**, 173 (2005).
- <sup>4</sup> H. Y. Hwang, *Science* **313**, 1895 (2006).
- <sup>5</sup> S. Thiel, G. Hammerl, A. Schmehl, C. W. Schneider, and J. Mannhart, *Science* **313**, 1942 (2006).