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Photoinduced insulator-to-metal transition in $ZnO/Mg_{0.15}Zn_{0.85}O$ heterostructures

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We report on the persistent photoconductivity accompanied with a steep insulator-to-metal transition at low temperatures in $ZnO/Mg_{0.15}Zn_{0.85}O$ heterostructures. The photoexcited electrons were confined in the ZnO adjacent to the $Mg_{0.15}Zn_{0.85}O$ to form a two-dimensional electron gas (2DEG). The electron density was controlled either by the power or number of ultraviolet laser pulses (266 nm wavelength) irradiated to the sample. The 2DEG exhibits Shubnikov-de Haas oscillation in magnetoresistance, whose oscillation periods coincide with the electron density evaluated by Hall effect measurements. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2841044]

Advanced epitaxial growth technology has substantially reduced crystalline defects and impurity concentrations in ZnO single crystalline thin films, leading to remarkably low residual electron density ($<1\times10^{16}$ cm^{-3}) and high mobility (>400 cm^2 V^{-1} s^{-1}).[1,2] By forming abrupt heterointerfaces composed of ZnO and $Mg_xZn_{1-x}O$, one can confine electrons into two-dimensional (2D) that exhibits quantum Hall effect (QHE) with high electron mobility exceeding 5000 cm^2 V^{-1} s^{-1} at 1 K.[3] In this system, the electron density (n) at the heterointerface was tuned by Mg contents and growth temperature (donor concentration). When we wish to discuss on the 2D transport phenomena at this heterointerface, it is important to develop a way for controlling n in such 2D electron gas (2DEG) in addition to the built-in structures. Application of electric field or photoirradiation are often used to control n in bulk semiconductors and heterostructures.[4–6] In fact, the photoconductivity has been investigated in modulation doped GaAs/AlGaAs heterostructures to control the 2DEG density and mobility.[7–11] In this letter, we present a photoirradiation technique applied to an insulating $ZnO/Mg_{0.15}Zn_{0.85}O$ heterostructure for investigating the transport properties of photoinduced electrons. We observe an insulator-to-metal transition (IMT) in the heterostructures by the irradiation of ultraviolet laser pulses. The photoconductivity persists in dark after photoirradiation and the metallic 2DEG state emerges with exhibiting Shubnikov–de Haas (SdH) oscillation.

$ZnO/Mg_{0.15}Zn_{0.85}O$ heterostructures were grown on $ScAlMgO_4$ (SCAM) substrates by pulsed-laser deposition in an oxygen pressure of 1×10^{-6} Torr.[1] First, 100-nm-thick $Mg_{0.15}Zn_{0.85}O$ layer was deposited and annealed *in situ* at 1000 °C in 1 mTorr of oxygen to make an atomically flat surface.[12] Then 20-nm-thick ZnO layer was deposited at 950 °C with observing intensity oscillation of reflection high-energy electron diffraction. The samples were processed into Hall bars with a geometry of 220×60 μm^2 by conventional photolithography and Ar ion milling, where Au/Ti layered metal electrodes were evaporated to form Ohmic contacts. The electrical properties were measured in physical properties measurement system (PPMS) (Quantum Design). We employed a pulsed ultraviolet laser (266 nm, 10 kHz) to shine the sample at 4 K in the PPMS (vacuum).

Temperature dependence of sheet resistance (ρ_{xx}) measured in dark is plotted in Fig. 1 by solid circles (●). Insulating behavior ($dR/dT<0$) was observed in the range of measurement temperature. When the laser pulses (0.2 W/cm^2, 1 s) were introduced from the surface of ZnO at 4 K, ρ_{xx} decreased by three orders of magnitude, as shown in the inset of Fig. 1. The recovery of ρ_{xx} was less than 0.1% for 25 min in dark, indicating persistent photoconcdutivity. After the photoirradiation, ρ_{xx} was measured while warming in dark, as shown by open circles (○), and we observed metallic behavior ($dR/dT>0$). At 400 K, ρ_{xx} merged with the original ρ_{xx} in insulating state. When the sample was cooled again from 400 K, ρ_{xx} traced the original curve, implying this IMT is reversible. Thermally activated relaxation

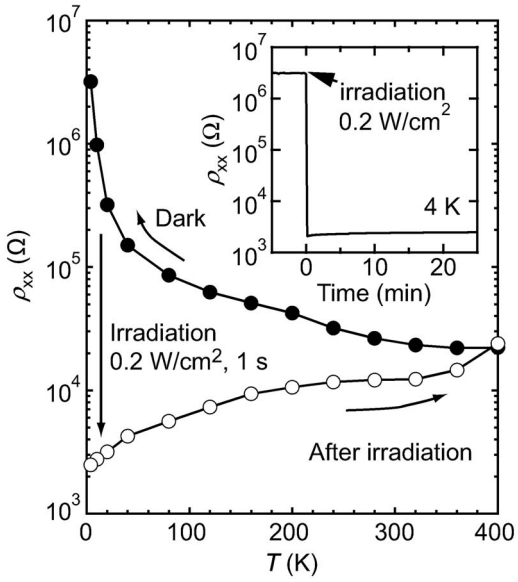

FIG. 1. Temperature dependence of ρ_{xx} in dark before (solid circles ●) and after (open circles ○) irradiation of 266 nm laser pulses. The inset shows the temporal variation of ρ_{xx} at 4 K after 1 s irradiation. The excitation conditions of the laser were a power of 0.2 W/cm^2, a repetition of 10 kHz, and a pulse width of 12 ns.

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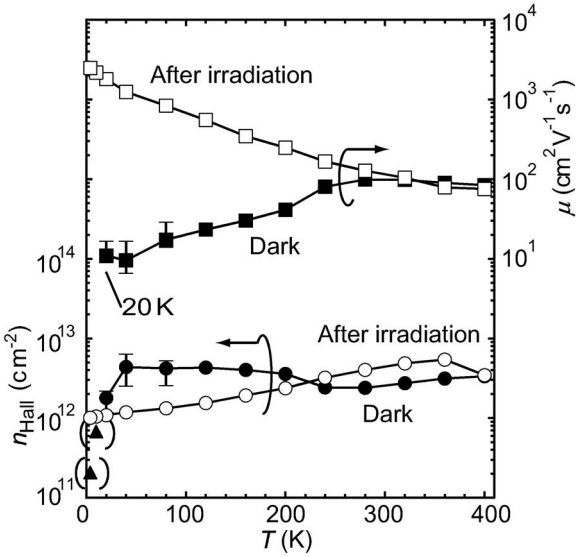

FIG. 2. Bottom: temperature dependence of n_{Hall} in dark before (solid circles ●) and after irradiation (open circles ○). Top: temperature dependence of μ in dark before (solid squares ■) and after irradiation (open squares □). The solid triangles (▲) show the calculated electron density at 10 and 4 K from the ρ_{xx} value with assuming $\mu=10\ \mathrm{cm^2\ V^{-1}\ s^{-1}}$.

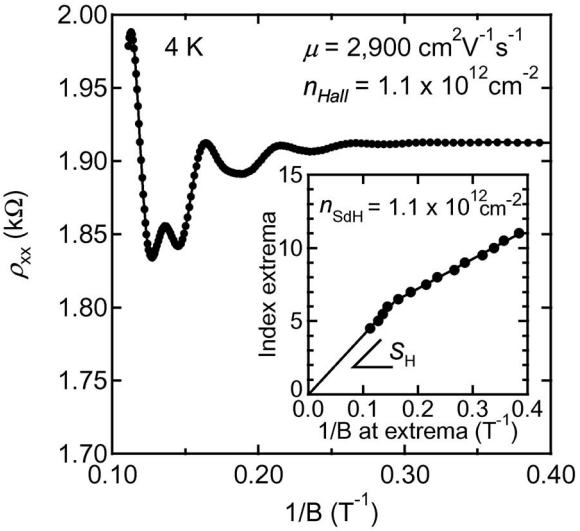

FIG. 3. Magnetoresistance measured at 4 K after photoirradiation. The inset shows indices of extrema in the oscillations plotted against inverse magnetic field.

was seen at moderately high temperature (>300 K). The ρ_{xx} in metallic state was gradually increased to recover ρ_{xx} in insulating state for hours.

During the measurement shown in Fig. 1, Hall effect measurements were also carried out. The temperature dependence of sheet electron density (n_{Hall}) and mobility (μ) is shown in Fig. 2. The n_{Hall} values in dark before and after irradiation were plotted by solid circles (●) and open circles (○), respectively, and the μ in dark before and after irradiation are presented by solid squares (■) and open squares (□), respectively. The n_{Hall} and μ in dark before irradiation were measured at low magnetic field (±1 T) in a temperature range from 400 to 20 K, where we could extract reliable values. Below 20 K, ρ_{xx} was too high to make reliable Hall effect measurement. Therefore, we plotted electron density by closed triangles (▲) with assuming that μ is 10 $\mathrm{cm^2\ V^{-1}\ s^{-1}}$ at 10 and 4 K. We repeated the measurement while warming up after photoirradiation at 4 K. As shown in Fig. 2, n_{Hall} before irradiation was almost constant until freezing out of carriers started below 40 K, but the μ decreased below 240 K probably due to disordered potential near the mobility edge. Upon irradiation at 4 K, μ showed drastic enhancement by more than two orders of magnitude and monotonically decreased with increasing temperature. The n_{Hall} and μ at 4 K after irradiation was 1×10^{12} $\mathrm{cm^{-2}}$ and 2500 $\mathrm{cm^2\ V^{-1}\ s^{-1}}$, respectively.

It is noted here that the major contribution to the enhancement of conductivity by photoirradiation comes from that of μ rather than n. Actually, n_{Hall} after irradiation was even lower than the values before irradiation below 220 K and gradually increased with increasing temperature. Although we do not have a good explanation for the butterfly-shape temperature variation of n_{Hall}, the observed temperature dependence of μ and n_{Hall} after irradiation is very similar to the one for a metallic sample exhibiting QHE,[3] suggesting the presence of 2DEG. If we assume the carrier was distributed in ZnO homogeneously, the three-dimensional (3D) electron density n ($\mathrm{cm^{-3}}$)=n_{Hall} ($\mathrm{cm^{-2}}$)/thickness (20 nm) would be 5×10^{17}–1×10^{18} $\mathrm{cm^{-3}}$, which is far less than 4×10^{18} $\mathrm{cm^{-3}}$ that is needed for 3D degeneracy at 300 K. If we calculate the effective 2DEG thickness using triangular-potential approximation, 10^{12} $\mathrm{cm^{-2}}$ electrons will be confined within 2 nm for the interface, resulting in 5×10^{18} $\mathrm{cm^{-3}}$ volume concentration higher than the threshold. The enhancement of μ by photoirradiation is not similar to the photoconductivity in ZnO bulk single crystals,[13,14] which is attributed to the enhancement of electron density. This difference would be directly related to the carrier dimensionality and, thus, scattering probability of the photoinduced electrons. Indeed, when we performed the same experiments for a ZnO single-layer film grown on SCAM substrate, we did not observe any IMT with photoirradiation (not shown).

The magnetoresistance was measured at 4 K for the photoirradiated metallic states and the result is shown in Fig. 3. We observed clear SdH oscillation that is one of the evidence for the presence of 2DEG at $\mathrm{ZnO/Mg_{0.15}Zn_{0.85}O}$ interface, because 3D carriers in ZnO have not been able to overcome the criteria of $\hbar\omega_c > k_BT$, where $\hbar$ is Planck's constant divided by 2π, ω_c is cyclotron frequency, k_B is Boltzmann's constant, and T is temperature. The inset shows indices of extrema plotted against inverse magnetic field (numbered with an increment of 0.5). The electron density contributing to the SdH oscillation (n_{SdH}) can be extracted from the high-field slope (S_H) by using the relation of $n_{\mathrm{SdH}}=eS_H/h$. The obtained n_{SdH} of 1.1×10^{12} $\mathrm{cm^{-2}}$ coincides with the n_{Hall}, indicating the photoinduced electrons are confined in lowest subband at the interface.

Next, we describe the controllability of n by varying the power or exposure time of laser irradiation. The inset of Fig. 4 shows n_{Hall} as a function of exposure time for subsequent irradiations at three different laser powers of 0.02 (●), 0.2(■), and 2(▲) $\mathrm{W/cm^2}$. The n_{Hall} could be continuously tuned up to 2.2×10^{12} $\mathrm{cm^{-2}}$ by varying laser power and exposure time. However, the increase of n_{Hall} was not linear with respect to the power of excitation laser. We estimate the quantum efficiency with an equation ($\eta=\Delta n/$ number of photons) and the value is found to decrease from 2% to below 0.1% with increasing the exposure time. The main panel of

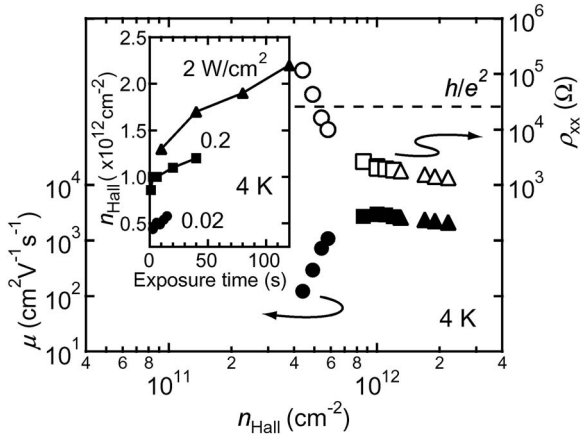

FIG. 4. The μ (solid symbols) and ρ_{xx} (open symbols) as a function of n_{Hall} measured at 4 K after light irradiation with three different power (0.02, 0.2, and 2 W/cm^2), each of which are depicted as solid circles (●), solid squares (■), and solid triangles (▲), respectively. The horizontal broken line at $\rho_{xx}=h/e^2$ indicates the quantum resistance. The inset shows exposure time dependence of n_{Hall} for various laser powers.

Fig. 4 shows the μ (solid symbols) and ρ_{xx} (open symbols) as a function of n_{Hall} at 4 K. The metallic states ($dR/dT>0$) were developed for the states with ρ_{xx} at 4 K lower than the quantum resistance (h/e^2) represented by broken line, which corresponds to n_{Hall} of about 6×10^{11} cm^{-2}. The steep enhancement in μ from 100 to above 1000 cm^2 V^{-1} s^{-1} coincides with the steep reduction in ρ_{xx}. The SdH oscillation was observable at the critical n_{Hall} required for developing metallic states. Above $n_{\mathrm{Hall}}\sim1\times10^{12}$ cm^{-2}, gradual decrease in μ is seen with increasing n_{Hall}. The positive magnetoresistance accompanied with SdH oscillation was observed when n_{Hall} was higher than 1.5×10^{12} cm^{-2}, probably due to the formation of parallel conduction 3D state in addition to the 2DEG.

Now, we discuss the origin of photoinduced carriers forming 2DEG state at the interface. Since the excitation photon energy of about 4.7 eV is greater than the bandgaps of ZnO (3.4 eV) and $Mg_{0.15}Zn_{0.85}O$ (3.6 eV), the photoinduced electrons should be generated both at ZnO and $Mg_{0.15}Zn_{0.85}O$ layers. The absorption coefficient of ZnO at 4.7 eV (266 nm) and at 4 K is about $\sim10^5$ cm^{-1} (Ref. 15) and, therefore, more than a half of laser power reach into $Mg_{0.15}Zn_{0.85}O$ through the 20-nm-thick of ZnO toplayer. Due to V-shaped band bending across the interface discussed in Ref. 3, photoinduced electrons in $Mg_{0.15}Zn_{0.85}O$ would be transformed to ZnO layer leaving photoinduced holes and ionized impurities in $Mg_{0.15}Zn_{0.85}O$. This results in spatial separation of the counter charges and is thought to be the origin of 2DEG leading to IMT and SdH oscillation.[9,10] If conducting electrons at the interface are not recombined with holes or captured in donor states in $Mg_{0.15}Zn_{0.85}O$, persistent state of 2DEG is developed. In this situation, the remote scattering by positive charges in $Mg_{0.15}Zn_{0.85}O$ would affect the mobile electrons at the interface. In low carrier density ($n_{\mathrm{Hall}}<10^{12}$ cm^{-2}) region, the enhancement of n promote the screening of those scattering centers in $Mg_{0.15}Zn_{0.85}O$, resulting in the improved mobility.[10] In high carrier density region ($n_{\mathrm{Hall}}>10^{12}$ cm^{-2}), the confinement of electrons also becomes strong with increasing the 2DEG density. Consequently, the enhancement of scattering probability arises from interface roughness as well as the remote scattering, resulting in the gradual reduction of μ shown in Fig. 4.[8]

In summary, persistent photoconductivity was observed upon ultraviolet photoirradiation for a $ZnO/Mg_{0.15}Zn_{0.85}O$ heterostructure. The IMT was observed due to steep enhancement of μ rather than the increase of n above a critical electron density. From observation of SdH oscillations, we have obtained strong evidence that the photoinduced electrons are confined at the heterointerface. We conclude from these results that the laser irradiation technique is an effective way for controlling the electron density in the present heterostructures.

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[1] A. Ohtomo and A. Tsukazaki, Semicond. Sci. Technol. **20**, S1 (2005).
[2] S. F. Chichibu, T. Onuma, M. Kubota, A. Uedono, T. Sota, A. Tsukazaki, A. Ohtomo, and M. Kawasaki, J. Appl. Phys. **99**, 093505 (2006).
[3] A. Tsukazaki, A. Ohtomo, T. Kita, Y. Ohno, H. Ohno, and M. Kawasaki, Science **315**, 1388 (2007).
[4] R. J. Nelson, Appl. Phys. Lett. **31**, 351 (1977).
[5] D. V. Lang and R. A. Logan, Phys. Rev. Lett. **39**, 635 (1977).
[6] M. I. Nathan, Solid-State Electron. **29**, 167 (1986).
[7] H. L. Stormer, R. Dingle, A. C. Gossard, W. Wiegmann, and M. D. Sturge, Solid State Commun. **29**, 705 (1979).
[8] H. L. Stormer, R. Dingle, A. C. Gossard, W. Wiegmann, and R. A. Logan, Inst. Phys. Conf. Ser. **43**, 557 (1979).
[9] H. L. Stormer, A. C. Gossard, W. Wiegmann, and K. Baldwin, Appl. Phys. Lett. **39**, 912 (1981).
[10] D. M. Collins, D. E. Mars, B. Fischer, and C. Kocot, J. Appl. Phys. **54**, 857 (1983).
[11] A. Kastalsky and J. C. M. Hwang, Solid State Commun. **51**, 317 (1984).
[12] A. Tsukazaki, A. Ohtomo, S. Yoshida, M. Kawasaki, C. H. Chia, T. Makino, Y. Segawa, T. Koida, S. F. Chichibu, and H. Koinuma, Appl. Phys. Lett. **83**, 2784 (2003).
[13] R. J. Collins and D. G. Thomas, Phys. Rev. **112**, 388 (1958).
[14] Y. Grinshpan, M. Nitzan, and Y. Goldstein, Phys. Rev. B **19**, 1098 (1979).
[15] O. Madelung, M. Schulz, and H. Weiss, *Numerical Data and Functional Relationships in Science and Technology, New Series*, Group III, Landolt-Börnstein Vol. 17, Subvol. b (Springer, Berlin, 1982).