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# Spatial distribution of two-dimensional electron gas in a ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterostructure probed with a conducting polymer Schottky contact

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A ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterostructure was characterized at  $T=2$  K through capacitance measurements with using a conducting polymer, poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS), as a Schottky contact. The Nyquist diagram, which is the trajectory curve of the complex impedance vector, appeared to be an excellent semicircular shape, implying that the PEDOT:PSS/ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O junction can be described with an equivalent single RC parallel circuit. Capacitance-voltage characteristics elucidate the existence of a two-dimensional electron gas, where  $10^{19}$  cm<sup>-3</sup> electrons are confined within 5 nm at the ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterointerface. © 2010 American Institute of Physics. [doi:10.1063/1.3309699]

Recent technological developments in ZnO thin film growth enabled us to realize reproducible acceptor doping, resulting in notable demonstration of ultraviolet (UV)/blue light-emission from  $p$ - $n$  homojunctions.<sup>1</sup> Besides its unique optical properties, superior electronic properties have been proven through the observation of quantum Hall effect for the high mobility two-dimensional electron gas (2DEG), which is spontaneously formed at ZnO/Mg<sub>x</sub>Zn<sub>1-x</sub>O heterointerfaces.<sup>2</sup> For improving the device performances of ZnO-based heterostructures, it is primarily important to elucidate the physical properties at ZnO/Mg<sub>x</sub>Zn<sub>1-x</sub>O heterointerfaces quantitatively. A metal/semiconductor Schottky junction is very useful for evaluating the fundamental electronic properties of semiconductors and heterostructures.<sup>3</sup> In particular, capacitance measurements on semiconductor heterostructures can extract important information such as a spatial carrier distribution and/or an energy band discontinuity at the interface.<sup>4</sup>

We have recently found an excellent Schottky contact for ZnO as follows: a well-known conducting polymer, poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS). Despite a simple fabrication technique based on a solution process, nearly ideal junction properties can be reproducibly obtained both for Zn-polar and O-polar ZnO surfaces.<sup>5,6</sup> In addition, by using such a significant feature of a PEDOT:PSS thin film that has a high internal transmittance of nearly 100% in a wide wavelength range from 250 to 800 nm, we have also demonstrated highly sensitive and color-selective UV light detectors.<sup>7,8</sup>

In this letter, we examine basic characteristics such as spatial distribution of 2DEG formed in a ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterostructure through capacitance measurements at  $T=2$  K by using PEDOT:PSS Schottky contact. The frequency dependence of the complex impedance revealed an equivalent circuit of the device as the parallel connection of

the depletion layer capacitance and resistance.<sup>9</sup>

Figure 1(a) shows a schematic device structure. A ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterostructure was prepared on an insulating ScAlMgO<sub>4</sub> substrate by pulsed laser deposition. The growth conditions are nominally the same as the previous report.<sup>2</sup> An atomically flat ZnO layer was formed on a high temperature annealed Mg<sub>0.2</sub>Zn<sub>0.8</sub>O buffer layer. In order to protect the O-polar ZnO surface from being dissolved in PEDOT:PSS acidic solution, the sample was first immersed for 1 min in a diluted solution of 3-aminopropyltriethoxysilane (APS) (0.1 mol/l) as previously reported.<sup>6</sup> PEDOT:PSS (H. C. Starck, Baytron PH500) was then spin-coated onto the specimen with a rotation speed of 4000 rpm followed by baking at 200 °C for 30 min, which yielded in a 50-nm-thick

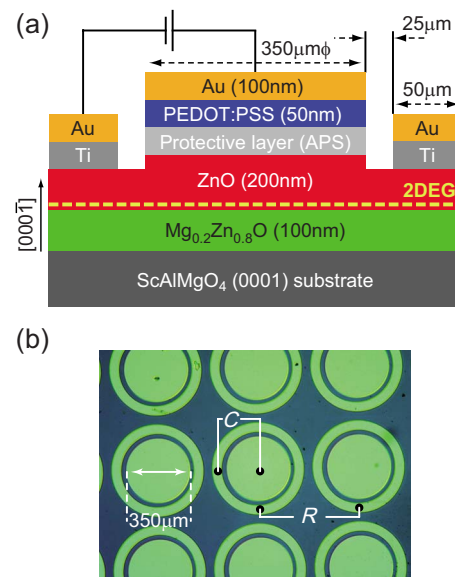


FIG. 1. (Color online) (a) A cross sectional schematic of the device. The diameters of circular mesa structure and the junction area were  $350\ \mu\text{m}$  and  $9.6 \times 10^{-4}\ \text{cm}^2$ , respectively. Broken straight line (yellow) represents the 2DEG at the ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterointerface. (b) The measurement configurations are shown on an optical microscope image of the devices.

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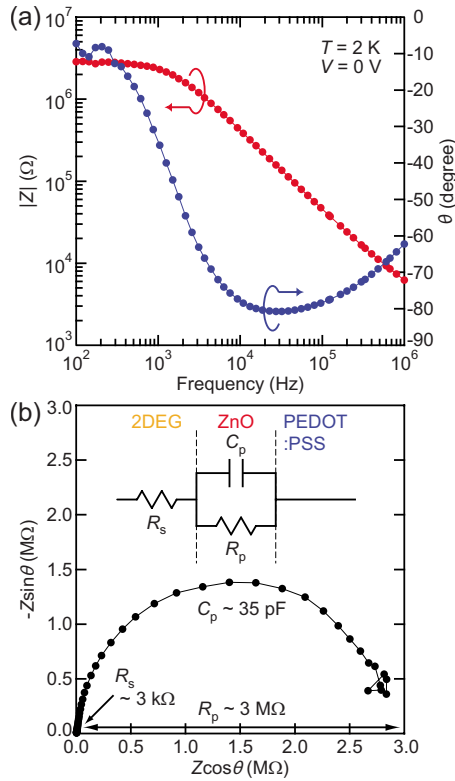


FIG. 2. (Color online) (a) The impedance spectrum (Bode diagram) of the device measured at  $T=2$  K under  $V=0$  V condition. The magnitude and the phase shift of the complex impedance,  $|Z|$  and  $\theta$ , were plotted against the measurement frequency ( $f$ ). (b) The trajectory curve of the Z vector (Nyquist diagram) calculated from the impedance spectrum. The equivalent electrical circuit is deduced as the inset. The values of the series resistance ( $R_s$ ), the parallel resistance ( $R_p$ ) and the parallel capacitance ( $C_p$ ) are also shown.

film. After the deposition of Au capping layer, conventional photolithography and Ar ion milling were used to form circular-shaped Schottky contacts and ring-shaped Ti/Au ohmic contacts as shown in Fig. 1(b). We made no annealing after the electrode deposition.

The resistance between two ohmic contacts ( $R$ ) showed metallic behavior against temperature (not shown). The roughly estimated  $R$  at  $T=2$  K was about 3 k $\Omega$ , which is consistent to the previous report,<sup>2</sup> implying the existence of 2DEG in a ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O heterostructure. Note that the voltage drop across the PEDOT:PSS layer can be neglected because the estimated longitudinal resistance of PEDOT:PSS layer at  $T=2$  K is as low as 0.1  $\Omega$ . The junction capacitance was examined from an impedance spectroscopy at  $T=2$  K by using an LCR meter (Agilent Technologies, 4284A).

Figure 2(a) shows the complex impedance ( $Z$ ) as a function of the measurement frequency ( $f$ ) (Bode diagram) measured at  $T=2$  K under zero-bias condition ( $V=0$  V). Assuming a series circuit of a resistance ( $R$ ), a capacitance ( $C$ ), and an inductance ( $L$ ), frequency dependence of  $Z$  is written as

$$Z = R + j \left( 2\pi fL - \frac{1}{2\pi fC} \right), \quad (1)$$

where  $j$  is the imaginary number. Therefore, a dominant component of a circuit can be extracted from a frequency dependence of  $Z$ . At low  $f$  below 1 kHz, the device showed almost constant  $|Z|$  of 3 M $\Omega$  and the phase shift ( $\theta$ ) of

$-10^\circ$ , corresponding to a resistive component of the device. By increasing  $f$  to 1 MHz,  $|Z|$  decreased monotonously and became almost inversely proportional to  $f$  above 10 kHz, corresponding to a capacitive component of the device. Meanwhile,  $\theta$  decreased with increasing  $f$  to minimum value of  $-80^\circ$  at around 40 kHz, and then gradually increased to  $-60^\circ$  at 1 MHz. This inverted bell-shape of  $\theta$ - $f$  curve is a signature of an electrical circuit with a single RC time constant.<sup>9-11</sup>

For further examination, we employ an equivalent circuit model. Figure 2(b) shows a trajectory curve of the Z vector (Nyquist diagram), in which the real and the imaginary parts of  $Z$  are plotted on x- and y-axes, respectively. The curve depicts a single semicircle, indicating the equivalent circuit of the device to be a single parallel circuit composed of a resistor ( $R_p$ ) and a capacitor ( $C_p$ ) with a series resistance ( $R_s$ ),<sup>9-11</sup> as shown in the inset of Fig. 2(b).  $R_p$  and  $C_p$  can be assigned to be a junction resistance and a capacitance of the depletion layer, respectively. From the diameter of the semicircle,  $R_p$  was estimated to be 3 M $\Omega$ . At the vertex of the semicircle ( $f=1.5$  kHz),  $C_p$  was determined to be 35 pF through a relation of  $R_p C_p = 1/(2\pi f)$ . The depletion layer width ( $x$ ) can be calculated from  $C_p$  with a relation of  $x = A\epsilon_s\epsilon_0/C_p$ , where  $A$  is the junction area,  $\epsilon_s$  is the relative permittivity and  $\epsilon_0$  is the vacuum permittivity. By assuming  $\epsilon_s=8.3$  (Ref. 12),  $x$  at  $V=0$  V was deduced to be 200 nm, which is close to the designed thickness of the ZnO layer. This correspondence suggests that PEDOT:PSS and 2DEG worked as parallel plates of a capacitor, and totally depleted ZnO layer acted as a dielectric at  $V=0$  V.  $R_s$  was assigned to be the sum of the series resistances of 2DEG, APS layer, and PEDOT:PSS layer.  $R_s$  was deduced to be 3 k $\Omega$ , which is consistent to the measured spread resistance of 2DEG ( $R$ ), implying negligible contribution of both APS and PEDOT:PSS layers on the electrical circuit. Good semicircular shape without distortion implies the formation of abrupt heterointerfaces both at PEDOT:PSS/ZnO and ZnO/Mg<sub>0.2</sub>Zn<sub>0.8</sub>O.<sup>9-11</sup> In addition, such a good semicircular shaped Nyquist diagram could be obtained even in a forward-biased region up to  $V=1$  V (not shown), although a certain dc current flows as shown in the inset of Fig. 3(a). This indicates that we can deduce the capacitance value correctly below  $V=1$  V. Under the situation, we can eliminate the possibility of multiple parallel and/or series RC circuits in the system.<sup>9-11</sup> Therefore, the voltage range in this study is limited below  $V=1$  V to ensure the validity of the capacitance value.

Figure 3(a) shows the  $C$ - $V$  (left axis) and the  $1/C^2$ - $V$  (right axis) characteristics measured at  $T=2$  K with  $f=40$  kHz.  $C$  decreases by changing  $V$  from +1 to -1.5 V with an increment of 10 mV, indicating the expansion of the depletion layer from ZnO layer into Mg<sub>0.2</sub>Zn<sub>0.8</sub>O layer. The slope of the  $1/C^2$ - $V$  curve determines the carrier concentration at the end of the depletion layer ( $x$ ) (Ref. 3), using a following relation of

$$n(x) = \frac{-2}{A^2 q \epsilon_s \epsilon_0} \frac{d(1/C^2)}{dV}. \quad (2)$$

Both  $C$  and  $1/C^2$  have plateau region, giving a strong evidence for the existence of a high carrier concentration layer at the corresponding depth. An increase of  $1/C^2$  with decreasing  $V$  from +1 to +0.8 V and below -0.5 V is due to

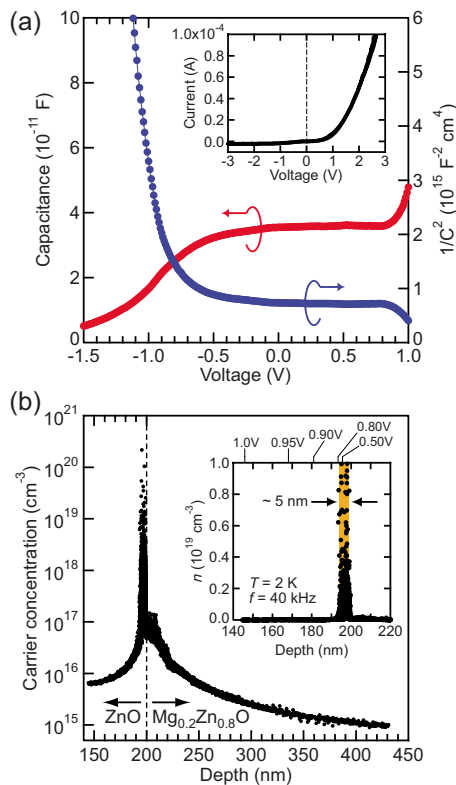


FIG. 3. (Color online) (a) A typical capacitance-voltage ( $C$ - $V$ ) characteristic measured at  $T=2$  K with  $f=40$  kHz. The corresponding  $1/C^2$ - $V$  characteristic was also shown. The inset shows a current-voltage ( $I$ - $V$ ) characteristic of the same device. (b) The depth profile of a representative device. The inset shows the same plot in linear scale. The bias voltages corresponding to the depth are shown on the top abscissa.

depletion of the ZnO layer and the  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  layer, respectively.

Figure 3(b) shows the depth profile of a representative device in logarithmic (main panel) and linear (inset) scales. These plots are gathered with all data from iterative  $V$  scans from  $+1$  to  $-1.5$  V over 50 times. An intense peak of 2DEG is clearly revealed at ZnO/ $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  heterointerface. The carrier concentration exceeds  $1 \times 10^{19} \text{ cm}^{-3}$  at the peak and these carriers are accumulated at the ZnO layer adjacent to the  $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  layer with a spreading width of 5 nm, which is consistent to the value estimated from a triangular poten-

tial approximation.<sup>13</sup> It is worth noting that the sheet carrier concentration is roughly estimated to be  $3 \times 10^{12} \text{ cm}^{-2}$  from the integration of depth profile, which is consistent with the typical values of ZnO/ $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  heterostructures evaluated by Hall coefficient and Shubnikov-de Haas oscillation period.<sup>2</sup>

In summary, we have characterized a ZnO/ $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$  heterostructure at  $T=2$  K through capacitance measurements by using PEDOT:PSS Schottky contact. The parallel RC equivalent circuit was identified from the frequency dependence of the complex impedance. The Nyquist diagram implied the formation of the ideal interfaces between constituent layers. The  $C$ - $V$  characteristics revealed the spatial distribution of 2DEG, which well agrees with the estimation based on a simple potential calculation. These results indicate that a PEDOT:PSS Schottky contact is very useful for making field-effect devices based on ZnO heterostructures.

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<sup>1</sup>A. Tsukazaki, A. Ohtomo, T. Onuma, M. Ohtani, T. Makino, M. Sumiya, K. Ohtani, S. F. Chichibu, S. Fuke, Y. Segawa, H. Ohno, H. Koinuma, and M. Kawasaki, *Nature Mater.* **4**, 42 (2005).

<sup>2</sup>A. Tsukazaki, A. Ohtomo, T. Kita, Y. Ohno, H. Ohno, and M. Kawasaki, *Science* **315**, 1388 (2007).

<sup>3</sup>S. M. Sze, *Physics of Semiconductor Devices*, 2nd ed. (Wiley, New York, 1981).

<sup>4</sup>H. Kroemer, W. Y. Chien, J. S. Harris, and D. D. Edwall, *Appl. Phys. Lett.* **36**, 295 (1980).

<sup>5</sup>M. Nakano, A. Tsukazaki, R. Y. Gunji, K. Ueno, A. Ohtomo, T. Fukumura, and M. Kawasaki, *Appl. Phys. Lett.* **91**, 142113 (2007).

<sup>6</sup>R. Y. Gunji, M. Nakano, A. Tsukazaki, A. Ohtomo, T. Fukumura, and M. Kawasaki, *Appl. Phys. Lett.* **93**, 012104 (2008).

<sup>7</sup>M. Nakano, T. Makino, A. Tsukazaki, K. Ueno, A. Ohtomo, T. Fukumura, H. Yuji, S. Akasaka, K. Tamura, K. Nakahara, T. Tanabe, A. Kamisawa, and M. Kawasaki, *Appl. Phys. Lett.* **93**, 123309 (2008).

<sup>8</sup>M. Nakano, T. Makino, A. Tsukazaki, K. Ueno, A. Ohtomo, T. Fukumura, H. Yuji, Y. Nishimoto, S. Akasaka, D. Takamizu, K. Nakahara, T. Tanabe, A. Kamisawa, and M. Kawasaki, *Appl. Phys. Express* **1**, 121201 (2008).

<sup>9</sup>E. Barsoukov and J. R. Macdonald, *Impedance Spectroscopy*, 2nd ed. (Wiley, New Jersey, 2005).

<sup>10</sup>I. M. Hodge, M. D. Ingram, and A. R. West, *J. Electroanal. Chem.* **74**, 125 (1976).

<sup>11</sup>J. T. S. Irvine, D. C. Sinclair, and A. R. West, *Adv. Mater.* **2**, 132 (1990).

<sup>12</sup>O. Madelung, M. Schulz, and H. Weiss, *Semiconductors: Physics of II-VI and I-VII Compounds, Semimagnetic Semiconductors*, Landolt-Börnstein, New Series, Group III Vol. 17, Pt. B (Springer, Berlin, 1982), p. 35.

<sup>13</sup>F. Stern, *Phys. Rev. B* **5**, 4891 (1972).