

Structural and resistance switching properties of ZnO/SrTiO₃/GaAs heterostructure grown by laser molecular beam epitaxy

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ZnO thin films were epitaxially grown on (001) GaAs substrate by laser molecular beam epitaxy with SrTiO₃ (STO) as a buffer layer. The interface properties of ZnO/GaAs heterostructure are greatly improved by inserting STO buffer layer. The interfacial effects on the transport and dielectric characteristics of the heterostructure have been investigated. The current-voltage characteristic of the heterostructure reveals an asymmetric and resistance switching behavior, exhibiting a temperature-dependent resistance hysteresis in the temperature range of 50–300 K. These measured properties could be attributed to the charge effect at the interface of the heterostructure. © 2010 American Institute of Physics. [doi:10.1063/1.3505136]

Efforts have been intensified in developing high-quality ZnO films and crystals on semiconductors for device applications.^{1–3} Epitaxial ZnO films have been reported to be fabricated on different substrates, such as GaN and sapphire,^{2–4} or introducing buffer layers.^{5,6} As it is known that optical and electrical properties of metal-oxide based films show high dependence upon interface structure, oxygen stoichiometry, and crystal orientation,^{4–7} those substrates served for depositing ZnO films have similar structure or close lattice parameter compared to that of ZnO. On the other hand, GaAs has widely been used in the microwave and optoelectronic industry due to its high electron mobility, direct band gap, and excellent optical characteristics.⁸ It could be expected that the integration of ZnO with GaAs will open the way to the fabrication of new devices with novel functionalities.^{9,10} However, it is a great challenge to epitaxial growth of ZnO on the cubic semiconductor due to lack of high quality, thermodynamically stable insulators that passivate the interface.^{10,11} Recently, low resistivity with low minority injection from polycrystalline ZnO on GaAs(001) substrate have been observed.¹² Ryu *et al.*¹³ reported the luminescence properties in polycrystal ZnO film on GaAs. High-quality self-textured p-ZnO has been synthesized on n-GaAs but the infrared electroluminescent emission characteristic of the heterostructure may suffer from the carrier injections.¹⁴ In our previous work, epitaxial growth of the SrTiO₃ (STO) was achieved directly on GaAs substrates by laser molecular beam epitaxy (LMBE) and subsequently the ferroelectric hysteresis loop in the STO buffered BaTiO₃(BTO)/GaAs was observed.^{15,16} In this work, we have fabricated the epitaxial ZnO thin film on GaAs via a STO buffer and investigated interfacial effects on the current-voltage (*I-V*) characteristics of the heterostructure.

ZnO/p-GaAs (001) heterostructure was grown via a STO buffer by O₂ flowing LMBE system with an operation wavelength of KrF ($\lambda=248$ nm, energy density of 6 J/cm²). In brief, first approximately 50 nm STO was deposited on

chemically cleaned p-GaAs (001) substrate at 600 °C. Subsequently, the substrate was heated to 650 °C and a 200 nm thick ZnO film was grown under O₂ partial pressure of 10 Pa. The film growth was monitored by *in situ* reflective high energy electron diffraction (RHEED) (Staib GmbH). Finally, Au metal electrodes with 0.2 mm in diameter were deposited for electrical measurements. The film structure was determined by x-ray diffraction (XRD). The measurement of *I-V* curve was performed by using an electrometer. The dielectric properties were measured by using a LCR meter.

Figure 1 shows RHEED patterns of the epitaxial feature of ZnO films grown on GaAs substrates using STO as a buffer layer. The diffraction spots of GaAs (001) in Fig. 1(a) gradually faded away as the STO started to grow. Figure 1(b) shows the diffraction pattern with 25 unit cells of STO (about 10 nm), indicating that the film was coherent at the onset of the STO growth. The clear elongated bright spots suggest that a well-defined single crystal STO layer forms on the GaAs substrate, which is consistent with the observation in Refs. 15 and 16. For the subsequent growth of the ZnO film on the STO buffered GaAs substrate, the diffraction pattern becomes weaker in contrast due to the interface transition from the perovskite to wurtzite structure. When the film thickness was greater than 3 nm, several Bragg-reflection spots gradually appeared as shown in Fig. 1(c), suggesting

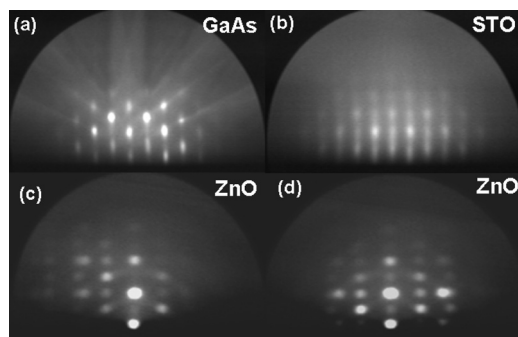


FIG. 1. RHEED patterns for heteroepitaxial growth of ZnO thin films on GaAs via STO buffer layer at various thickness: GaAs substrate (a), 10 nm STO (b), 5 nm ZnO (c), and 200 nm ZnO (d).

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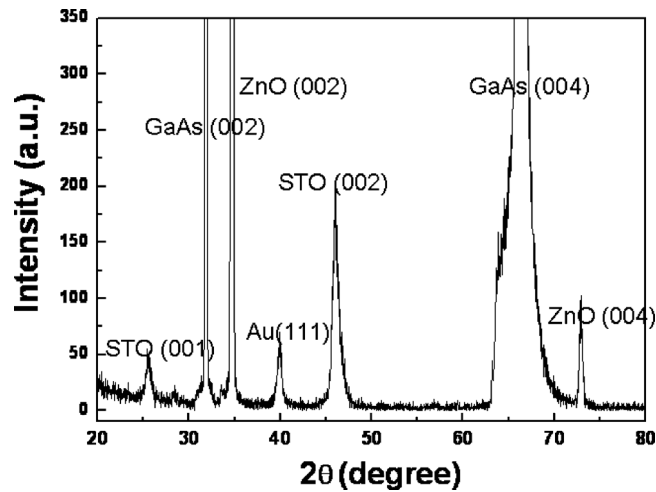


FIG. 2. XRD pattern of the ZnO/STO/GaAs heterostructure.

that the film grows with the island growth mode. With increasing ZnO film thickness further, no unidentifiable RHEED patterns were detected, and the quality of the diffraction pattern was enhanced as shown in Fig. 1(d), suggesting that the epitaxy and crystallinity of ZnO film remained satisfactory via a STO interfacial layer.

Figure 2 shows a typical θ - 2θ XRD scan of the ZnO/STO/GaAs heterostructure. Only the (000 l) peaks of the ZnO appear in the diffraction pattern, indicating that the ZnO is basically c-axis oriented growth on the STO buffered GaAs substrate. To further evaluate the in-plane alignment of the film, Φ scan was performed (not shown here), which reveals that the ZnO film is epitaxially grown on the (001) surface of STO and has an in-plane film-substrate orientation relationship of $[1\bar{1}00]\text{ZnO}/[1\bar{1}0]\text{STO}/[100]\text{GaAs}$, which is consistent with the results from RHEED observation.

Figure 3(a) shows a schematic diagram of the Au/ZnO/STO/GaAs heterostructure. The heterostructure exhibits a variation in dielectric constant as a function of frequency at room temperature. The dielectric constant can be calculated to be 64 at 1 kHz. As shown in Fig. 3(a), the experimental dielectric constant value of heterostructure was smaller compared to that of STO bulk and decreased significantly with the increase in measuring frequency. The reduced dielectric constant may be attributed to the ZnO thin film with a small dielectric constant. Figure 3(b) shows the capacitance-voltage curve at high frequency (1 MHz) between -6 and 6 V. There exhibits almost “zero” hysteresis in this C - V curve with the maximum shift less than 50 mV. The extremely low hysteresis, along with the sharp transition from depletion and accumulation, reveals the high quality of crystalline and interface properties of the heterostructure.¹⁷

Figure 4 shows a typical I - V curve of the heterostructure at room temperature, indicating a resistance switching characteristic. The I - V curve reveals a strong asymmetric behavior. When the bias voltage was swept as $-5 \rightarrow 0 \rightarrow +5 \rightarrow 0 \rightarrow -5$ V, a forward-to-reverse bias ratio of 125 was obtained at the voltage of ± 4 V. It is apparent that the heterostructure exhibits a high resistance state when sweeping from 0 to $+5$ V, and a low resistance state when sweeping from $+5$ to 0 V. We have also measured hysteresis I - V characteristic for different bias voltage (not show here). All I - V curves exhibit a similar hysteresis property. It is interesting to note that the

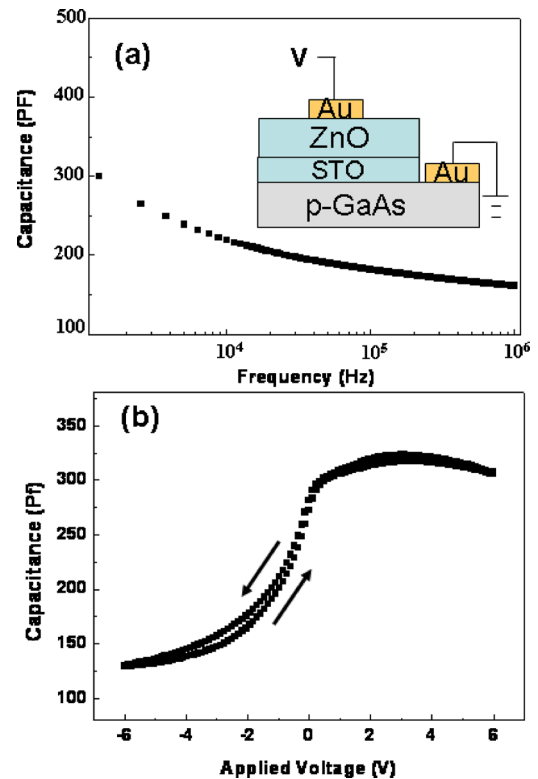


FIG. 3. (Color online) The capacitance of the Au/ZnO/STO/GaAs heterostructure as a function of frequency (a), the inset shows the schematic of the Au/ZnO/STO/GaAs heterostructure and the capacitance-voltage curve of the heterostructure under the bias voltage of 6 V at the frequency of 1 MHz (b). The arrows denoted the direction of the bias sweeping.

junction shows larger I - V loop with higher bias voltage. In previous studies, reversible electroresistive (ER) switching effect was found in some oxide systems.¹⁸ Recently, a lot of research has been focused on the switching properties of ZnO thin films.^{19,20} Although the mechanism is not well understood yet, the interface effect should play an important role in controlling the ER switching and other electric properties. For instance, in ZnO/BTO/ZnO heterostructure, the switching behaviors have been explained by coupling of the ferroelectric BTO and piezoelectric ZnO interface charges.²⁰ In our case, the origin of the switching behavior seems to be quite complicated. According to the previous studies on ZnO/GaAs heterostructure,^{13,14} a diodelike rectifying I - V

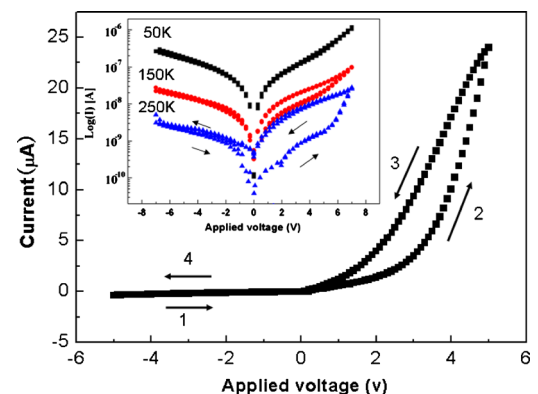


FIG. 4. (Color online) Current-voltage property under the bias voltage of 5 V of the heterostructure at room temperature. The arrows indicate the bias voltage was swept as $-5 \rightarrow 0 \rightarrow +5 \rightarrow 0 \rightarrow -5$ V. The inset shows the current-voltage properties (in log scale) at various temperatures.

characteristic without the switching behavior has been observed. The similar results have also been founded in STO/GaAs and other STO/GaAs related heterostructures.^{16,17} As a consequence, we consider that the switching behavior in the ZnO/STO/GaAs heterostructure could be attributed to the interface contact with a Schottky barrier between ZnO/STO interface. It is known that interface properties in STO based heterostructure could differ quite considerably.^{21,22} The observation may be related to a scenario of charging effect at the interface through Fowler–Nordheim tunneling to defect states; i.e., the acceptor trapping states present in the Schottky barrier because of impurities and structural defects.²³

In order to get more insights into the current transport mechanism in the Au/ZnO/STO/GaAs heterostructure, temperature-dependent *I-V* measurements have been carried out in the temperature range of 50–300 K, as shown in the inset of Fig. 4. While the temperature decreases, the hysteresis (in logarithmic scale) window is getting smaller. At 50 K, the hysteresis disappears while the diodelike behavior is persistent. This result is quite different compared with the temperature-dependent switching behavior in the ZnO/BTO/ZnO system,²⁰ which is caused by the disappearance of the charge coupling between the ferroelectric and the piezoelectric interface above the Curie temperature of BTO due to the phase transition from its tetragonal ferroelectric to cubic paraelectric phase in the BTO material. By contrast, in our case, the disappearance of the hysteresis can be considered as disappearance of the Schottky barrier at a low temperature. This is somewhat similar to the temperature-dependent *I-V* curve in other systems of metal-oxide-semiconductor structures, such as Al/HfO₂/SiO₂/p-Si,²⁴ TaN/ZrO₂/SiO₂/p-Si,²⁵ etc. These temperature-dependent *I-V* behaviors always show similar performances by inducing an interfacial layer. Therefore, these reversible ER switching effects in our heterostructure could also be considered as the interface contact with a Schottky barrier between ZnO and the STO buffer layer.

In summary, the epitaxial growth of the ZnO thin film on GaAs has been achieved using STO as a buffer layer. The interface properties of ZnO/GaAs heterostructure are greatly improved by inserting a 50 nm thick STO buffer layer. The behavior of temperature-dependent reversible resistance switching was observed in the heterostructure.

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