Review Article

Two-Dimensional Electron Gas at SrTiO_3 -Based Oxide Heterostructures via Atomic Layer Deposition

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Two-dimensional electron gas (2DEG) at an oxide interface has been attracting considerable attention for physics research and nanoelectronic applications. Early studies reported the formation of 2DEG at semiconductor interfaces (e.g., $\text{AlGaAs}/\text{GaAs}$ heterostructures) with interesting electrical properties such as high electron mobility. Besides 2DEG formation at semiconductor junctions, 2DEG was realized at the interface of an oxide heterostructure such as the $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) heterojunction. The origin of 2DEG was attributed to the well-known “polar catastrophe” mechanism in oxide heterostructures, which consist of an epitaxial LAO layer on a single crystalline STO substrate among proposed mechanisms. Recently, it was reported that the creation of 2DEG was achieved using the atomic layer deposition (ALD) technique, which opens new functionality of ALD in emerging nanoelectronics. This review is focused on the origin of 2DEG at oxide heterostructures using the ALD process. In particular, it addresses the origin of 2DEG at oxide interfaces based on an alternative mechanism (i.e., oxygen vacancies).

1. Introduction of 2DEG

Two-dimensional electron gas (2DEG) has been observed at the interface of III–V semiconductor heterostructures, such as the $\text{AlGaAs}/\text{GaAs}$ interface, with unusual high electron mobility [1–3]. Besides the $\text{AlGaAs}/\text{GaAs}$ heterointerface, nitride-based semiconductor heterojunctions such as the AlGaN/GaN heterostructure have been extensively explored owing to its high mobility of electrons at the interface. Their electrons are able to move freely along the interface direction while confined within a few nanometers at the interface (interface normal direction) [4, 5]. The origin of 2DEG at semiconductor interfaces was due to the formation of a quantum well at the junction. This 2DEG at semiconductor interfaces is greatly useful in optoelectronic devices as well as transistors for power electronics and radiofrequency (RF) application because 2DEG plays a key role in the operation of field effect transistors (FETs). This makes oxide heterojunctions highly valuable [6]. In the meantime, significant interest has returned to 2DEG since its observation at the oxide heterojunctions (i.e., $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO)) with high electron density up to $10^{14}/\text{cm}^2$, which is about two

orders of magnitude higher in electron density than that of semiconductor heterojunctions (Figure 1) [7]. Interestingly, it was revealed that a large number of electrons were confined at the heterojunction of LAO and STO layers. Thus, significant experimental researches had been performed as well as theoretical approaches [8–27]. Recently, interesting properties of 2DEG at oxide heterostructures have been explored regarding superconductivity and ferromagnetism [28–36]. The 2DEG at oxide heterojunctions is potentially applicable for high-performance transparent transistors due to the high density of electrons and high transparency in the visible range [11].

The LAO/STO heterostructure was the model system in the oxide heterostructure for 2DEG. The LAO layers were usually grown by pulsed laser deposition (PLD) or molecular beam epitaxy (MBE) on single crystalline STO substrates at the typical growth temperature of $>600^\circ\text{C}$. The LAO overlayer had to be grown epitaxially on the STO substrates to achieve abrupt and sharp interfaces because the origin of 2DEG via the polar catastrophe mechanism requires an atomically perfect interface based on polar and nonpolar characteristics of LAO and STO layers [7, 8, 10, 13, 22]. STO has a cubic ABO_3 -type perovskite crystal structure while LAO shows

FIGURE 1: Structural illustration of 2DEG at the interface of LAO/STO heterostructure.

a rhombohedral crystal structure (distorted pseudocubic perovskite) [37]. The perovskite such as LAO consisted of LAO and AlO_2 planes along [001] direction, where each LAO and AlO_2 plane exhibited +1 ($\text{La}^{3+}\text{O}_2^{2-}$) and -1 ($\text{Al}^{3+}\text{O}_2^{4-}$) charges, respectively, considering valence states of La^{3+} , Al^{3+} , and O^{2-} in the LAO. However, STO is composed of neutral SrO ($\text{Sr}^{2+}\text{O}_2^{2-}$) and TiO_2 ($\text{Ti}^{4+}\text{O}_2^{4-}$) planes because of valence states of Sr^{+2} , Ti^{4+} , and O^{2-} , respectively. In the case of heterointerface of LAO/STO, an electrostatic potential across the interface diverged with the LAO film thickness due to polar discontinuity [10]. It was proposed that if half of the electrons per unit cell transfer from LAO to STO, the overall structure is neutral and the electrostatic potential no longer diverged across the interface, which is a well-known polar catastrophe mechanism in the origin of 2DEG [10]. In this electrostatic charge consideration at the atomic scale, the TiO_2 termination was essential for the STO because TiO_2 and SrO species are mixed on the surface of commercially available STO substrates. The TiO_2 -termination step has been reported elsewhere [38, 39].

Although many studies have indicated polar catastrophe as the plausible mechanism for the origin of 2DEG at oxide heterostructures, this has been under debate. Other possible origins of an electron source, such as oxygen vacancies and La interdiffusion, could be involved in the origin of 2DEG at oxide heterostructures [14–18, 40–42]. Experimentally, it has been reported that when an STO substrate was reduced in certain growth conditions (such as high growth temperature with low oxygen partial pressure) to create oxygen vacancies, these vacancies provided electrons in the LAO/STO heterostructure [14]. It should be noted that the origin of carriers at LAO/STO heterostructures strongly depends on the growth conditions of the LAO/STO heterostructures, which might have induced the debates in the origin of 2DEG among researchers [43].

2. ALD in Emerging Applications

The atomic layer deposition (ALD) process is operated by alternative injections of a metal precursor and oxygen (or sulfur and hydrogen) source for the growth of oxides (or sulfides and metal thin films) while the purge step is carried out between the metal precursor and oxygen source injection steps. The metal precursor injection-purge-oxygen source

injection-purge steps consist of one deposition cycle that can be repeated until the target thickness is achieved. The history of the ALD technique began in the 1970s to grow functional films such as ZnS layers for flat-panel display applications [44–48]. The ALD technique has received great attention because it was considered as a key process for the growth of functional thin films, which is suitable for mass-production in the semiconductor industry. Thus, a rapid progress was accomplished in the development of semiconductor devices such as dynamic random access memories (DRAM) in the 1990s. Besides, for the conventional semiconductor applications, the ALD technique has recently been spotlighted in emerging nanoelectronics, optoelectronics, and sensor applications, because it enabled a high-quality deposition of extremely thin films in complicated structures at a low temperature ($<300^\circ\text{C}$). On one side, the ALD technique has a potential to be used for emerging nanoelectronic and optoelectronic devices because of the compatibility to state-of-the-art semiconductor technology; on the other side, it is useful for new applications, such as “double patterning” to complement current lithography technology, in which thin films grown by ALD determined a pattern size [49].

3. ALD for 2DEG at Amorphous LAO/STO Heterostructures

It has been reported that a conductive channel was formed using an amorphous LAO overlayer by the PLD process at an LAO/STO heterostructure; thus, the amorphous LAO layer has emerged as a new overlayer for the origin of 2DEG on STO substrates [17]. The LAO layer was grown by PLD at room temperature to make the amorphous layer on the STO substrate. In that case, a conductive channel was created with a high density of electrons (up to $\sim 10^{14}/\text{cm}^2$) at the amorphous LAO/STO interface. This was similar to that of an epitaxial LAO/STO interface using PLD, in which oxygen vacancies contributed to the generation of carriers in the amorphous LAO/STO heterostructure. Interestingly, STO/STO and YSZ (yttria-stabilized zirconia)/STO heterostructures (which have amorphous overlayers) also exhibited high conductance at the interface while heterostructures with the amorphous overlayers grown on LAO substrates showed an insulating interface. This indicates that the STO substrates (i.e., oxygen vacancies in the STO substrates) are important for 2DEG creation [17].

Recently, it was revealed that an amorphous LAO layer was capable of creating 2DEG via the ALD process [50]. The origin of 2DEG via the ALD technique has great significance in the practical application of 2DEG because the ALD process is suitable for the demands of industries such as those in the semiconductor field. Therefore, it opens new possibilities in the application of 2DEG at oxide heterostructures. Besides the capability of ALD for the practical application of 2DEG, realization of 2DEG via ALD could offer direct experimental evidence in finding a 2DEG origin [50]. This review addresses the origin of 2DEG at oxide heterostructures via ALD.

In order to create 2DEG using ALD, an amorphous LAO layer was grown by ALD at 300°C on a TiO_2 -terminated STO

FIGURE 2: (a) Sheet resistance, carrier density, and (b) carrier mobility of amorphous LAO/STO heterostructures as a function of LAO overlayer thickness (reproduced from [50] with permission).

substrate with very smooth surface morphology (rms roughness of 0.2 nm) [50]. Lanthanum tris($\text{N,N}'$ -diisopropylformamidinate) and trimethylaluminum (TMA) were used as La and Al precursors, respectively. The oxygen source was H_2O vapor. The amorphous nature of the LAO layer was confirmed by X-ray diffraction (XRD) and a transmission electron microscope (TEM); thus, the polar catastrophe mechanism was excluded in the amorphous LAO/STO heterostructure for the origin of 2DEG. A TiO_2 -terminated STO substrate was also necessary for 2DEG creation in the amorphous LAO/STO heterostructure. The resistance of 2DEG without the TiO_2 -termination of the STO substrate was 2~3 orders of magnitude higher than that with the TiO_2 -termination step. Figure 2(a) shows the sheet resistance and carrier density of LAO/STO heterostructures as a function of LAO layer thickness, which represents the metal-insulator transition across the critical LAO thickness of ~2.5 nm. The insulating interface of LAO/STO changed to a conductive interface with increasing LAO overlayer thickness. A substantial number ($\sim 10^{13}/\text{cm}^2$) of electrons were created above the critical LAO thickness while the carrier mobility remained constant as shown in Figure 2(b) ($4\text{--}5 \text{ cm}^2/\text{Vs}$). The type of carrier was electron as confirmed by the Hall voltage, and above the critical LAO layer thickness, the carrier density approached $\sim 10^{13}/\text{cm}^2$ irrespective of the LAO layer thickness. It was confirmed by angle-resolved X-ray photoelectron spectroscopy (angle-resolved XPS) that the depth of 2DEG was ~2 nm from the interface in the STO side. These 2DEG properties are almost identical to those of epitaxial LAO/STO heterostructures, which proved the ALD technique useful in emerging new applications of 2DEG. The growth of an amorphous oxide by ALD is a technically mature process in the semiconductor industry and encourages the actual application of 2DEG in nanoelectronics and optoelectronics.

It was found that 2DEG creation using an amorphous LAO overlayer was achieved owing to the key role of TMA

(the Al precursor) in the LAO ALD process for the reduction of the STO surface (discussed in Section 5). This aspect allowed a wider window of the La:Al composition (40~60 at% of La:Al ratio) for the creation of 2DEG using an amorphous LAO overlayer compared to that of an epitaxial LAO/STO interface which needs stoichiometric LAO overlayer.

4. ALD for 2DEG at Epitaxial LAO/STO Heterostructures

The ALD process has been used to grow epitaxial thin films on compound semiconductors such as GaAs to obtain a low density of interface traps for next-generation high-performance FETs beyond silicon transistors. High dielectric constant (high- k) thin films, such as $\text{La}_x\text{Lu}_y\text{O}$ and $\text{La}_x\text{Y}_y\text{O}$, were grown epitaxially on GaAs substrates using ALD, resulting in significantly decreased interface trap densities [51, 52]. The epitaxial growth of $\text{La}_x\text{Lu}_y\text{O}$ and $\text{La}_x\text{Y}_y\text{O}$ thin films was achieved by lattice parameter engineering between the high- k thin films and the GaAs substrate. For the $\text{La}_x\text{Y}_y\text{O}$ case, composition (La:Y ratio) was controlled precisely for high-quality heteroepitaxy on the GaAs (111) substrate. Likewise, for heteroepitaxy on GaAs, growing an epitaxial LAO layer on the STO substrate using ALD has been tried to create 2DEG via the polar catastrophe mechanism. One encouraging report was the growth of an epitaxial LAO layer on the TiO_2 -terminated STO substrate by ALD, in which tris(isopropylcyclopentadienyl)lanthanum and TMA were used for La and Al precursors, respectively [53]. The composition of the LAO layer was controlled by the cycle ratio of La precursor pulse and Al precursor pulse. The LAO layer grown by ALD on the TiO_2 -terminated STO substrate was amorphous at a growth temperature of 300°C . Thus, the amorphous LAO/STO heterostructure was annealed at 900°C to crystallize the amorphous LAO layer. The crystalline structure

was analyzed by TEM with a selective area diffraction pattern, showing an epitaxial interface between the LAO layer and STO substrate with a (100) plane of LAO on a (100) STO substrate. The epitaxial growth of the LAO layer on the STO substrate by ALD is an encouraging result. However, it is not clear whether the epitaxial interface was achieved through the whole area of the sample, and the interface of the LAO/STO heterostructure might be intermixed because of the high annealing temperature (900°C), which can induce La/Sr interdiffusion at the interface of LAO and STO. This suggests that *in situ* epitaxy of the LAO layer would be preferable to *ex situ* epitaxy using the annealing process. The epitaxial LAO/STO heterostructure using ALD showed an approximately three orders of magnitude lower current (15–4000 pA at 10 V) than that of other reported LAO/STO structures. Thus, the resistance of the epitaxial LAO/STO interface was approximately three orders of magnitude higher than that of typical LAO/STO heterostructures. Moreover, the I–V curve was not linear, which indicates ohmic contact was not formed because of an insufficient 2DEG creation. Such a low current might result from insufficient epitaxial layer quality and low oxygen vacancies due to the high temperature annealing in oxygen ambient. It is anticipated that 2DEG would be created by the polar catastrophe mechanism if *in situ* epitaxy of the LAO layer on the STO substrate is achieved during the ALD process.

5. ALD for 2DEG at Amorphous-Al₂O₃/STO Heterostructures

One important observation was the creation of 2DEG using an amorphous Al₂O₃ overlayer (i.e., an Al₂O₃/STO heterojunction that did not include La atoms) grown by ALD [50]. The amorphous Al₂O₃ overlayer was grown by ALD on a TiO₂-terminated STO substrate at 300°C using TMA and H₂O. It was the first report that a binary oxide was capable of 2DEG generation on STO-based oxide heterostructures by means of the ALD process. So far, a perovskite-structured overlayer such as an LAO layer has been used to create 2DEG. However, the grown Al₂O₃ layer on the STO substrate was amorphous. This observation offered significant advances for finding direct evidences in STO-based oxide heterostructures through the alternative 2DEG creation mechanism (i.e., oxygen vacancies). In the Al₂O₃/STO heterojunction, the polar catastrophe mechanism was excluded because the overlayer was amorphous, whereas the polar catastrophe mechanism is valid only at an epitaxial interface with an atomically abrupt and sharp interface. In addition, the Al₂O₃ layer was not perovskite-structured material, like the LAO layer having a polar nature of the atomic plane. Furthermore, the La interdiffusion issue was neglected in the 2DEG origin at the Al₂O₃/STO heterostructure due to the absence of La components in the Al₂O₃/STO heterostructure. Therefore, it was possible to conclude that oxygen vacancies are the plausible origin of 2DEG in oxide heterostructures, especially in the case of amorphous overlayers using the ALD technique.

The oxygen vacancies were generated on the surface of STO substrates during the growth of an Al₂O₃ overlayer

FIGURE 3: Illustration of 2DEG creation mechanism at amorphous Al₂O₃/STO heterostructure via ALD (reproduced from [54] with permission from the Royal Society of Chemistry).

by ALD which were detected by X-ray photoelectron spectroscopy (XPS). In principle, the valence state of the Ti ion is +4 in the defect-free STO substrate; however, the valence state of the Ti ion changes to +3 if oxygen vacancies exist in the STO substrate [50, 54]. XPS analysis indicated a Ti³⁺ peak after the growth of the Al₂O₃ layer by ALD on the STO substrate, suggesting that substantial oxygen vacancies were formed on the STO surface during the growth of the Al₂O₃ overlayer by ALD. It is well known that the generation of oxygen vacancies donates free electrons in the STO, implying that the oxygen vacancies are a primary electron source in Al₂O₃/STO heterostructures. Equation (1) shows one possible reaction scheme to create oxygen vacancies in the STO substrate by using TMA during ALD of the Al₂O₃ layer [50]:

The change of Gibbs free energy in this reaction was $\Delta G = -1063 \text{ kJ/mol}$ at 573 K, indicating the transition from Ti⁴⁺ to Ti³⁺ by TMA was thermodynamically favorable [50, 55]. It has been proposed that the oxygen ions (in STO) diffuse out to oxidize the reactive species which is adsorbed on the STO surface during the growth of LAO layer by PLD process [17]. Like the PLD process, it is likely that oxygen ions diffuse outward to oxidize the TMA which is adsorbed on the STO substrate during the ALD process. Although the thermodynamic criteria implied the proposed reaction could occur, the reaction rate was determined by STO reduction kinetics. Figure 3 illustrates how the adsorbed TMA molecules generated oxygen vacancies in the STO surface during the growth of the Al₂O₃ overlayer by ALD at 300°C. It was supposed that the reduction of STO occurred at the TMA injection step during ALD, and the reaction was kinetically limited because the density of electrons changed

FIGURE 4: Illustration of the critical thickness on conductivity, (a) below and (b) above the critical thickness of Al_2O_3 /STO heterostructures (reproduced from [54] with permission from the Royal Society of Chemistry).

as a function of ALD temperature (i.e., the electron density decreased with decreasing growth temperatures). In order to confirm the role of TMA, any ALD process that involved the Al_2O_3 deposition sequence using TMA was attempted (e.g., a YAlO_x overlayer, which can be deposited by intermixing Y_2O_3 and Al_2O_3 layers by ALD). Interestingly, 2DEG was also created by the growth of the amorphous YAlO_x layer at 300°C . However, the creation of 2DEG was not achieved by deposition of only an amorphous Y_2O_3 layer, without an Al_2O_3 layer, on the STO substrate [50]. In addition, 2DEG was created even though the Y composition in the YAO layer exceeded 62% (Y:Al ratio), which was far from the stoichiometric YAO composition. This suggested that the polar catastrophe mechanism was not involved in these amorphous overlayer/STO heterostructures. In the meantime, it was proposed that the generation of oxygen vacancies was limited by the increased overlayer thickness. While oxygen atoms should diffuse out to create oxygen vacancies in the STO substrate, oxygen transmission through the overlayer became difficult as the overlayer thickness increased. Thus, the amount of Y_2O_3 in the YAO layer did not affect 2DEG creation because the Y_2O_3 layer is highly transparent to oxygen transmission compared Al_2O_3 [56]. The Al_2O_3 layer has a significantly lower oxygen transmission rate; thus, the oxygen vacancy generation rate was limited by the thickness of the Al_2O_3 layer [54].

One curious point was the existence of the critical thickness in the amorphous overlayer/STO heterostructures. For the epitaxial interface of the LAO/STO heterostructure, it was reported that the polar catastrophe mechanism required at least four LAO overlayer unit cells for electron transfer from the overlayer to STO surface; however, it cannot be valid on the amorphous overlayer [11]. Interestingly, it turned out that the oxygen-deficient area under the grown Al_2O_3 layer widened as the Al_2O_3 layer became thicker as shown in Figure 4; therefore, it should be sufficiently connected to achieve a macroscopically conductive heterostructure with such a kind of Hall measurement [54]. This aspect is the origin of critical overlayer thickness on the macroscopic conductivity of the Al_2O_3 /STO heterostructure. However,

this point does not mean that a conducting path does not exist under the critical thickness of the Al_2O_3 overlayer, but that the conductive area (by oxygen vacancies) needs to be connected sufficiently with adjacent conducting areas for the conducting path generation [54].

6. ALD for 2DEG at Epitaxial Al_2O_3 /STO Heterostructures

Chen et al. reported 2DEG at an epitaxial Al_2O_3 /STO interface with a spinel $\gamma\text{-Al}_2\text{O}_3$ phase on the STO substrate by PLD at 700°C (Figure 5) [57]. Epitaxial growth of the Al_2O_3 layer was possible because the oxygen sublattice of the $\gamma\text{-Al}_2\text{O}_3$ phase was matched with that of the STO (001) substrate (lattice mismatch of 1.2%). Interestingly, 2DEG was realized at the epitaxial Al_2O_3 /STO interface with a highly enhanced electron mobility of $\sim 140,000\text{ cm}^2/\text{Vs}$ at 2 K. The electron mobility of $140,000\text{ cm}^2/\text{Vs}$ was a record high in the 2DEG system with STO-based oxides heterostructures. In addition, quantum oscillation characteristics were observed due to a quantum confinement effect. Shubnikov-de Haas (SdH) oscillation (resistance oscillation by a magnetic field) was apparent with angle-dependent measurements, which further confirmed a two-dimensional system in the Al_2O_3 /STO heterostructure. Although epitaxial growth of the Al_2O_3 layer was achieved on the STO substrate, the polar catastrophe mechanism does not fit the epitaxial Al_2O_3 /STO interface because the $\gamma\text{-Al}_2\text{O}_3$ layer is not composed of a polar plane, unlike the LAO layer. The origin of 2DEG at the $\gamma\text{-Al}_2\text{O}_3$ /STO heterostructure was explained by oxygen vacancies in the STO substrate, which is consistent with that of the amorphous Al_2O_3 /STO heterostructure. This implies that epitaxial growth is not always necessary for 2DEG creation [50].

More recently, an epitaxial interface was reported at a $\gamma\text{-Al}_2\text{O}_3$ /STO heterostructure via ALD [58]. The $\gamma\text{-Al}_2\text{O}_3$ phase was realized on the STO (001) substrate by ALD using a typical ALD scheme for Al_2O_3 deposition. This was the same ALD scheme, using TMA and H_2O , as reported by Lee et al. [50]. However, the ALD process was performed at 345°C , which is a slightly higher growth temperature for Al_2O_3

FIGURE 5: Lattice match between $\gamma\text{-Al}_2\text{O}_3$ and STO (001) substrate, indicating possible epitaxial interface (reproduced from [57] with permission).

deposition than that (300°C) used by Lee et al. Figure 6 shows the TEM image of the Al_2O_3 layer grown on TiO_2 -terminated STO (001) substrates by ALD at 345°C (Figures 6(a), 6(c), and 6(d)) and 300°C (Figure 6(b)). A high-quality epitaxial $\gamma\text{-Al}_2\text{O}_3$ phase was obtained at the growth temperature of 345°C , as confirmed by TEM images and the selective area electron diffraction pattern. Figure 6(d) exhibits discrete points indicating a single crystalline phase of the Al_2O_3 layer. However, Al_2O_3 film grown at 300°C also exhibited a high degree of crystallinity, which is not consistent with Lee et al.'s previous report that an amorphous Al_2O_3 layer was obtained at 300°C [42, 46]. The observation of Ti^{3+} at the growth temperature of 300°C was not consistent between two reports [50, 54]. These contradictions might be related to temperature calibration or reoxidation of the samples by H_2O during ALD or air exposure after ALD. Amorphous Al_2O_3 layer growth was observed at 200°C in both studies. The depth of 2DEG

in the epitaxial $\gamma\text{-Al}_2\text{O}_3/\text{STO}$ heterostructure was estimated to be 1.7 nm, which is similar to that of the amorphous $\text{Al}_2\text{O}_3/\text{STO}$ heterostructure (~ 2 nm).

In both studies (amorphous Al_2O_3 and $\gamma\text{-Al}_2\text{O}_3$), it was revealed that the origin of 2DEG using an amorphous Al_2O_3 or $\gamma\text{-Al}_2\text{O}_3$ layer was due to oxygen vacancies. This fact implies that the phase of the Al_2O_3 overlayer has nothing to do with the origin of 2DEG at $\text{Al}_2\text{O}_3/\text{STO}$ heterostructures; that is, the oxygen vacancies were the source of electrons regardless of the phase of the Al_2O_3 overlayer. Eventually, the epitaxial layer was not always required at $\text{Al}_2\text{O}_3/\text{STO}$ heterostructures for the creation of 2DEG.

7. Conclusion

This review addressed the origin of 2DEG at LAO/STO and $\text{Al}_2\text{O}_3/\text{STO}$ heterostructures via ALD depending on

FIGURE 6: Epitaxial growth of γ -Al₂O₃ on TiO₂-terminated STO substrate via ALD at (a) 2.1 nm thick Al₂O₃ at 345°C, (b) 2.5 nm thick Al₂O₃ at 300°C, (c) 27 nm thick Al₂O₃ at 345°C, and (d) selective area electron diffraction of 27 nm thick Al₂O₃ at 345°C (reproduced from [58] with permission).

the phase of the overlayer. It also discussed the amorphous overlayer as well as the epitaxial overlayer deposited by ALD for 2DEG creation. With the ALD technique, 2DEG was generated during the growth of amorphous and epitaxial overlayer on the STO substrates. Regardless of the phase of ALD-grown overlayers on STO substrates, the oxygen vacancies on STO surface were the primary origin of the electron source for 2DEG. In any case of 2DEG at STO-based oxide heterostructures using ALD, the electron densities were in the range of $10^{13} \sim 10^{14}/\text{cm}^2$. These electron densities are comparable with those of the oxide heterostructures using other deposition techniques such as PLD and MBE, and the electron mobilities are also similar ($\sim 5 \text{ cm}^2/\text{Vs}$) because the electrons may move via Ti 3d subbands in the STO.

To date, the ALD technique has been used successfully for various applications to meet the demand in nanoelectronics and optoelectronics. In particular, the ALD technique was capable of creating 2DEG at oxide heterostructures such as LAO/STO and Al₂O₃/STO heterostructures. The origin of 2DEG at oxide heterostructures using the ALD technique can be applied to the mass-production in the semiconductor industry owing to the process compatibility and low cost compared to those of PLD and MBE processes. Thus, the

potential application of 2DEG at oxide heterostructures can provide promising opportunities in emerging nanoelectronics and optoelectronics.

Competing Interests

The author declares that there are no conflicts of interest regarding the publication of this paper.

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