Heterojunction oxide thin-film transistors

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Semiconducting Metal Oxide Thin-Film Transistors

Authors:

Hendrik Faber (<https://orcid.org/0000-0002-4918-2365>)

Emre Yarali

Emre Yengel (<https://orcid.org/0000-0001-7208-4803>)

Thomas D. Anthopoulos* (<http://orcid.org/0000-0002-0978-8813>)

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1 Introduction

Metal oxide (MO) semiconductors find widespread application in a variety of (opto)electronic devices due to a multitude of beneficial properties. Transparency and high electron mobility together with decent environmental stability, non-toxicity, and a range of possible deposition schemes have made them an excellent material class for thin-film transistor (TFTs) applications of all kinds, with specific emphasis on display technologies for (consumer) electronics (1-3). Target requirements in this field are a high charge carrier mobility μ_{FE} , which allows small pixel sizes and fast refresh rates that can cope even with demanding applications *e.g.* in virtual reality displays, as well as good stability during continuous use cases over extended time periods. Both requirements should ideally be met using materials and fabrication methods that are cost-effective and easily applicable over large areas. Aside from chasing this goal via optimizing individual materials, through composition and process control, another approach that has gained attention in the recent years is to make use of active channels composed of layers of two or more different MO materials (4-6). These heterojunction (HJ) films can lead to remarkable device properties when integrated into TFTs, allowing the improvement of both targets, μ_{FE} and bias stability, simultaneously. In many cases, the HJ TFTs achieve better results than either of the individual MO films is capable of.

The proposed reasons and mechanisms for an improved performance that will be discussed in the course of this chapter may vary from case to case. Among them are the creation of a two dimensional electron gas (2DEG) at the HJ (7), the combination of positive properties of individual MO layers while mitigating their drawbacks (8), the control of chemical composition, stoichiometry and morphology as well as interplays between them.

When referring to metal oxide HJ films there is a number of terms that can be found in literature, *e.g.* bilayer metal oxide (BMO) (9), dual active layer (DAL) (10, 11), double-active layer (12) stacked active layer

(SAL) (13), or (quasi)-superlattice (14-16). Especially the latter two expressions hint at the fact that the number of layers in a HJ film is not limited to two, but that multiple layers of different MOs can be stacked together. In addition to multilayers, this chapter also discusses reports of homojunction films where the respective layers feature nominally the same material (*e.g.* IGZO) which were processed in a way to create distinct properties per layer (17-21).

Heterojunction films are created by sequential deposition of the individual components. In the literature, different naming conventions can be found when describing the sequence of a bilayer film. While describing a film stack of $\text{In}_2\text{O}_3/\text{ZnO}$, for one research group this refers to the sequence starting from the substrate, i.e. an In_2O_3 is being deposited first onto the substrate, followed by ZnO layer on top. Other authors, however, may use the same term ($\text{In}_2\text{O}_3/\text{ZnO}$) to refer to the opposite stacking direction, starting with the material on the very top. To avoid possible confusion, throughout this chapter we will be using the first naming convention that describes layers according to the sequence in which they are deposited, as depicted schematically in Figure 1. Another way to refer to individual films is by the names front channel and back channel, whereas front channel is the film directly at the interface with the dielectric and the back channel is the film facing away from the dielectric, typically in contact with the environment or a passivation layer for the bottom-gate top-contact device architecture. Due to the multitude of different metal oxides and their combinations in ternary or higher compound materials, abbreviation such as IGZO for indium gallium zinc oxide will be used hereafter. In these abbreviations, the following letter-element combinations will be employed: I for Indium, G for Gallium, Z for Zinc, T for Tin, A for Aluminium, H for Hafnium, O for Oxide. Among the material pairings within the HJs, there can be found junctions between insulator/insulator (22), semiconductor/semiconductor (14, 23, 24), or semiconductor/insulator (15, 25-28).

In the course of this chapter we will first take a look at the underlying mechanisms for the improved devices performances caused by HJ metal oxide (HJMO) films, as well as typical material combination and

deposition techniques used to process the HJs (section 2). Next, section 3 is providing a general overview of HJMO TFTs and their field-effect mobility μ_{FE} as the main figure of merit. It then goes on describing examples how specific parameters, such as film thickness ratio or film stacking sequence in HJ and multilayer TFTs, can have an impact on the device characteristics. Finally, section 4 showcases the effects that HJ channels have to offer in regards to bias stress stability followed by the conclusion and outlook section 5.

Figure 1: Naming convention for heterojunction film stacks as part of a TFT. The expression MO1/MO2 refers to metal oxide 1 being the first film to be deposited onto the substrate or dielectric and metal oxide 2 being the top layer.

2 Mechanisms and materials for heterojunction TFTs

2.1 2DEG formation

A single semiconductor channel material will always be bound to its inherent limitations when it comes to charge carrier mobility, but the combination of two or more materials allows the exploitation of interface effects that enable new transport properties as well as mobility values that surpass those measured for the individual HJ components. Joining two materials with differently sized bandgaps and a large enough conduction band offset ΔE_c can lead to the formation of a potential well at the heterointerface in which electrons are confined and can form a 2-dimensional electron gas (2DEG). This

phenomenon has been shown in epitaxially grown HJs based on *e.g.* GaAs/AlGaAs (29) or AlGaN/GaN (30). An additional electron transfer into the potential well can be initiated by a process called modulation doping (MOD) where the barrier layer is intentionally doped. This results in an increased electron density in the 2DEG and, importantly, it physically separates the ionized donors from the charge carriers. As a consequence, the effects of impurity scattering are all but eliminated which allows the carrier mobility to reach larger value than that measured for the bulk semiconductors that form the HJ (29, 31).

For oxide based materials, 2DEG formation has been reported *e.g.* for heterojunctions based on the insulating perovskite oxides SrTiO₃ and LaAlO₃ (22) as well as for ZnO/ZnMgO (32-34). While the alternating AO and BO₂ planes that define the perovskite structure are charge neutral for the case of SrTiO₃ (Sr²⁺O²⁻ and Ti⁴⁺(O²⁻)₂) they either carry one positive or negative charge for LaAlO₃ (La³⁺O²⁻ and Al³⁺(O²⁻)₂). This leaves the HJ of SrTiO₃/LaAlO₃ with either half an extra electron or hole per unit cell that can be transferred across the interface where it is spatially confined (4). Ohtomo *et al.* investigated SrTiO₃/LaAlO₃ HJ deposited by pulsed laser deposition (PLD) (22). They reported that the junction formed by Al³⁺(O²⁻)₂/ Sr²⁺O²⁻ remains an insulator, however, if the junction is formed by La³⁺O²⁻ and Ti⁴⁺(O²⁻)₂ it shows n-type conductivity and a high Hall mobility μ_H above 10,000 cm²/Vs measured at 2 K. The polarity discontinuation at the interface was proposed as the main mechanism for the electron transfer and 2DEG creation.

The formation of a 2DEG in the ZnO/ZnMgO system was reported as early as 2003. Eda Hiro *et al.* used PLD to grow a ZnMgO/ZnO HJ on a sapphire substrate and as a result found a high sheet carrier concentration of $8.8 \times 10^{12} \text{ cm}^{-2}$ and a clear bump in carrier density at the location of interface, revealed by capacitance-voltage (C-V) carrier profiling (32). A high room temperature (RT) Hall mobility μ_H of 130 cm²/Vs was determined, which increased further for reduced temperatures down to 100 K then remained stable and featured a value of 360 cm²/Vs at 1.85 K. This temperature dependant behaviour was indicative of suppressed ionized impurity scattering as compared to a single ZnO layer, whose mobility fell drastically

for $T < 100$ K. Additionally, clear Shubnikov-de Haas (SdH) oscillations were observed in magnetoresistance measurements at 1.85 K. In 2004, Koike *et al.* realized a double heterojunction of ZnMgO/ZnO/ZnMgO, grown by molecular beam epitaxy (MBE) on sapphire (35). A deep potential well was formed as a result of the large conduction band discontinuity ΔE_c of 0.9 eV between both oxides. The lattice mismatch between ZnO and ZnMgO lead to a polarization induced high sheet charge and the accumulation of free electrons to form the 2DEG at the interface. Hall effect measurements deduced an electron density of $1.2 \times 10^{13} \text{ cm}^{-2}$ as well as μ_H of 170 and 400 cm^2/Vs at RT and 77 K, respectively. Tampo *et al.* altered the MBE conditions to successfully grow the HJ on Zn polar faces of ZnO where it was previously only possible to use the O polar surface (23, 36). This resulted in a boost in μ_H with a value of 250 cm^2/Vs at RT and up to 2750 cm^2/Vs at 4 K. Further refinement of the Mg content later improved μ_H to 319 cm^2/Vs at RT (7). In work by Nakano *et al.* (37), the authors report a ZnO/ZnMgO HJ grown by MBE on Zn faced (0001) ZnO substrates, where a conductive polymer is spin cast on top of the stack to act as a Schottky contact. This allowed field-controlled measurements of Hall mobility with a value up to 21,000 cm^2/Vs at 2 K. This value was further increased in studies by Tzukasaki *et al.* who were able to observe fractional quantum Hall effect in a ZnO/ZnMgO HJ and reached μ_H of 180,000 cm^2/Vs at 0.06 K (33) as well as a report by Falson *et al.*, who measured a maximum μ_H of 700,000 cm^2/Vs at 0.5 K in films with a very dilute Mg concentration (38).

Collectively, the early investigations pointed out a number of properties that are exemplary for the formation of a 2DEG. Among those are a high conduction band offset ΔE_c to confine the electrons at the HJ (Figure 2a), typically a high sheet carrier density on the order of 10^{12} - 10^{13} cm^{-2} which is largely independent of temperature, and a clear peak in the cross-sectional carrier profile at the location of the HJ (Figure 2b,c). Additionally, the importance of a high quality and flat interface of the HJ was highlighted (7, 39). Further, the mobility was found to be increasing with decreasing temperature in contrast to a

mobility decline at low temperatures for a single layer ZnO. Finally, HJs that formed a 2DEG showed SdH oscillations at low temperature magnetoresistance measurements (inset in Figure 2c).

The first transistors using a MO HJ with a 2DEG in the ZnO/ZnMgO system were demonstrated between 2005 and 2007 by Koike *et al.* (40) and Sasa *et al.* (41, 42). All cases used MBE to grow the channel materials on top of sapphire substrates and employed a top-gate device architecture with MgO, Al₂O₃ or HfO₂ as gate dielectrics and mobilities μ_{FE} between 62 and 240 cm²/Vs were achieved. Despite the exceptionally high mobility values, a drawback for practical applications of these structures was the low drain current on/off ratio that stayed below 10³.

Figure 2: Properties observed for 2DEGs HJ films. a) Band diagram of ITZO/IGZO HJ with proposed 2DEG formation. The band offset makes it favourable for electrons to accumulate at the HJ interface. Reproduced with permission. (43) Copyright 2018, John Wiley and Sons. b) Electron carrier concentration across a ZnO/ZnMgO HJ cross-section in the direction from the top surface towards the substrate. A clear spike in carrier concentration indicates the location of the 2DEG at the interface of the HJ. Reprinted from (7), with the permission of AIP Publishing. c) Temperature dependent mobility and sheet carrier concentration

for a ZnO/ZnMgO system. The inset shows SdH oscillations in magnetoresistance measurements at 0.5 K. Reprinted from (7), with the permission of AIP Publishing. d) Plot of square root of drain current vs gate voltage for solution processed ZnO, In_2O_3 and bilayer $\text{In}_2\text{O}_3/\text{ZnO}$ TFTs. The HJ device features improved performance over both individual MO films. Reproduced with permission. (44) Copyright 2020, John Wiley and Sons.

In 2010, the first experiments were carried out to confirm that 2DEGs could be obtained with other deposition methods that did not rely on epitaxial growth on special substrates. Chin *et al.* used RF sputtering to fabricate HJs of ZnO/ZnMgO on glass substrates and despite the polycrystalline nature of the films observed 2DEG characteristics, such as a high sheet carrier density, which increased to a maximum around $1 \times 10^{14} \text{ cm}^{-2}$ with increasing Mg content due to increased polarization (45). Additionally, both the carrier density and the Hall mobility were found to be approximately temperature independent. It was argued that the high carrier density of the 2DEG could successfully screen the grain boundary potential (46) but that improvements in surface roughness were required to increase the only moderate μ_{H} of $39.4 \text{ cm}^2/\text{Vs}$. The same approach of sputtered ZnO/ZnMgO was integrated by Cheng *et al.* to produce top-gated HJ TFTs with Al_2O_3 dielectric and achieved a high μ_{FE} of $84.2 \text{ cm}^2/\text{Vs}$ while maintaining a large on/off ratio $> 10^6$, indicating the practical applicability of polycrystalline metal oxide HJs forming 2DEGs (47). Similarly, Remashan *et al.* used metal organic chemical vapor deposition (MOCVD) to grow polycrystalline ZnO on top of a ZnMgO layer and reported an increased μ_{FE} of the HJ TFT of $9.1 \text{ cm}^2/\text{Vs}$ compared to a single layer of ZnO ($2.3 \text{ cm}^2/\text{Vs}$) (48).

The two dimensional nature of electrons was also described in materials systems other than crystalline ZnO/ZnMgO, for example for a thin polycrystalline ITO layer on top of amorphous IGZO or in superlattices based on ZnO and Al_2O_3 (15, 49). Taniguchi *et al.* described a type 2 band offset for their sputter deposited IGZO/ITO heterojunction that favors electron transport from the ITO conduction band to that of IGZO and the 2D nature of electrons was assigned based on temperature dependent Hall effect measurements (49). Resulting HJ TFTs achieved a μ_{FE} of $20 \text{ cm}^2/\text{Vs}$ compared to $7.5 \text{ cm}^2/\text{Vs}$ for a single layer of IGZO. On the other hand, Ahn *et al.* produced semiconductor/dielectric superlattices by atomic layer deposition (ALD)

of ZnO/Al₂O₃ at moderate 200 °C on common Si/SiO₂ substrates (15). ZnO films grown on alumina showed improved crystallinity and preferred c-axis orientation compared to ZnO grown on the bare substrate. Together with the 2D confinement of electrons in the ZnO layers due to the large Al₂O₃ band gap, this superlattice channel showed enhanced mobility (27.8 cm²/Vs) and on/off ratio (10⁹) compared to ZnO only devices, as well as improved bias stress behavior.

After 2DEG formation had been observed even for imperfect, polycrystalline vacuum deposited films, the question arose if similar effects could also be realized with solution processing methods. To this end, Lin *et al.* developed In₂O₃/ZnO HJs based on ultrathin (< 10 nm) films processed via spin coating of precursors and subsequent annealing at moderate temperatures below 200°C (14). Due to an offset in Fermi level and conduction band, an electron transfer from ZnO to In₂O₃ was proposed, where available energy levels are quantized because of the low film thickness, leading to a quasi 2D-electron-gas-like system at the MO HJ. The bilayer structure was extended by adding thin layers of equally solution processed Ga₂O₃ to form a quasi superlattice (QSL) system consisting of In₂O₃/Ga₂O₃/ZnO/Ga₂O₃/In₂O₃. The addition of Ga₂O₃ served the purpose to enhance confinement and interface planarity as well as for trap passivation. This QSL setup exhibited high μ_{FE} of 30 and 40 cm²/Vs. using a SiO₂ or using a Al₂O₃/ZrO₂ bilayer dielectric, respectively. The concept around using discrete solution processed In₂O₃ and ZnO bilayers was further developed by the same research group, exploring a new solution-based deposition technique (spray pyrolysis) (50), the effects of high temperature annealing up to 600 °C (51), photonic curing as alternative method for the rapid precursor conversion (44, 52), the impact of modulation doping (MOD) through Li-doped ZnO films (53), the variation in stacking sequence within multilayer junctions (16) and the incorporation of ZnO nanoparticles and an interlayer of polystyrene for additional trap passivation (54). Several properties that resemble or hint at a 2DEG system were observed repeatedly in these studies, including a spike in electron concentration at the location of the heterojunction (via C-V carrier profiling), a change towards a temperature independent μ_{FE} and a shift in transport properties from a trap limited

conduction (TLC) to a percolation conduction (PC) (14, 16, 44, 50). Additionally, the mobility of the various HJ structures reached consistently high values between $\approx 20 - 50 \text{ cm}^2/\text{Vs}$, and in all cases was significantly enhanced when compared to TFTs based on either of the individual MO components (as depicted in Figure 2d). This synergetic effect demonstrates the unique properties that can be achieved by two-dimensional systems engineered at metal oxide heterojunctions.

Other solution processed MO bilayer systems for that 2DEG formation was reported include *e.g.* the combination of ITZO/IGZO (43), $\text{In}_2\text{O}_3/\text{AlInO}$ (55), or a homojunction of undoped and polymer doped In_2O_3 (56). Lee et al. first investigated the properties of planar junctions depending on the bottom ITZO layer thickness, and then created a corrugated stripe structure of ITZO/IGZO bilayers with alternating thin/thick IZO bottom layers in the stripes running perpendicular to the channel (43). Here, the thick-IZO/IGZO parts were identified to enhance μ_{FE} , while the thin-IZO/IGZO parts were able to maintain a low off-current level. This combination resulted in TFTs with high mobility up to $51 \text{ cm}^2/\text{Vs}$ in combination with an on/off ratio $> 10^8$. In the study by Chen *et al.* the authors used the polymer polyethylenimine (PEI) to dope the back channel In_2O_3 on top of an undoped In_2O_3 front channel (56). The resulting energy offsets gave rise to electron confinement and 2DEG formation. Transistor performance based on $\text{In}_2\text{O}_3/\text{In}_2\text{O}_3:\text{PEI}$ surpassed that of single layer In_2O_3 devices and reached mobilities $> 10 \text{ cm}^2/\text{Vs}$ and as high as $30 \text{ cm}^2/\text{Vs}$ using SiO_2 or a high-k ZrO_2 dielectric, respectively.

2.2 Dual active layers and other mechanisms

While HJ layer stacks that form a 2DEG typically lead to TFTs in which the bilayer is showing improved characteristics (*e.g.* higher mobility and currents, improved bias stability) compared to either of the individual films, another widely used approach to work with layered MOs is to balance out properties of either components (6, 9, 21, 57). In this case, a thin but highly conductive MO with high charge carrier

density N_e and mobility μ_{FE} , typically deposited as the front channel, serves as the main transport layer. By itself, the drain current for this film may be difficult to modulate through the gate voltage resulting in either a negative V_{th} or even the device not being able to fully turning off. In order to achieve successful gate modulation, a second and more resistive MO with smaller mobility is used, mostly as the back channel material. Compared to devices based on the front channel material alone, this bilayer combination can have the drawback to reduce the drain current in the on-state slightly but it comes with the substantial benefit that it is now possible to fully turn-off the device. For this combination, the back channel takes control over the turn-on behavior and V_{th} , while the front channel still delivers reasonably high currents and mobility values. Because both layers play their distinctive part in the assembly, the term dual active layer is often used to describe them. An example of this behavior is shown in the transfer characteristics in Figure 3, where a rather conductive In_2O_3 is combined with layer of IGO and the combined solution processed In_2O_3 /IGO bilayer film shares the off-current with IGO yet still delivers an increased I_D in the on-state (8).

Figure 3: a) Schematic cross-section transfer characteristics of TFTs based on single layer In_2O_3 and IGO as well as their combination in an In_2O_3 /IGO dual active layer. The bilayer combines beneficial attributes from both components, such as the higher mobility and on-current of the front channel and the low off-current of the back channel material. Reprinted with permission from (8). Copyright 2013 American Chemical Society.

The approach to use a high mobility transport / high N_e transport layer with a more stable film to control off-current and V_{th} has been pursued with a number of metal oxide combinations and different deposition techniques. For sputter deposited HJs this includes *e.g.* ITO/ZTO (58), IZO/HIZO (59, 60), IZO/IGZO (61) or IGZO:H/IGZO (21). Similarly, solution based dual layer HJ TFTs have been reported based on *e.g.* IZO/AlZO (62, 63), In_2O_3 /IGO (8), or also indium-free composition of SnO/ZnO (64).

Other reasons to employ a bilayer HJ include the control and improvement of chemical composition, stoichiometry or morphology. For example, as reported for sputtered as well as solution processed back channels of tin containing materials, i.e. IZO/ZTO (65), IGZO/ITO (10) or IGZO/ZTO (11), the diffusion of Sn^{4+} into the respective bottom layer could lead to improvements in device performance. The tin cations served as n-dopants by substituting for In^{3+} , Ga^{3+} or Zn^{2+} , thus increasing the carrier concentration in the channel.

Metal oxide films deposited by various solution processes may still possess a considerable degree of porosity as well as pinholes which are detrimental to an efficient charge transport. Consequently, the subsequent application of a second MO layer has the chance to fill up gaps in the bottom layer and thus help to improve the channel properties. This beneficial effect was observed for spin cast ZTO films deposited on top of IGZO (10, 11), as well as for inkjet printed ZTO/ In_2O_3 bilayers in the study by Lee *et al.*, who reported film densification through pore filling and passivation as well as reduced surface roughness leading to improved μ_{FE} in devices (12).

In a rare example of p-type metal oxide HJs, Al-Jawhari *et al.* conducted a study on SnO/ Cu_2O TFTs, where the Cu_2O capping layer could be used to help control the SnO stoichiometry (66). Depending on the oxygen pressure during sputter deposition, the layer thickness ratios, and annealing temperatures between 140 and 200 °C, the resulting TFTs were either found to exhibit n-type behavior, p-type behavior or no

functionality. With the correct parameter set, an improved p-type mobility of $0.66 \text{ cm}^2/\text{Vs}$ could be established compared to $0.23 \text{ cm}^2/\text{Vs}$ for a single film of SnO.

It is worth noting that the various possible origins for improved device characteristics of HJ TFTs described here in section 2 do not have to be seen as isolated mechanisms. Rather, a combination of several of them acting simultaneously may be responsible and the exact root cause determination for enhanced charge transport properties may not always be possible.

3. TFTs based on bilayer heterojunctions and multilayer stacks

3.1 Overview

Before looking at a few specific examples how the properties of HJMO TFTs can be tailored, Figure 4 is showing an overview of mobility values that were reported to date, with respect to the deposition methods and maximum substrate temperature (Figure 4a) as well as the year of publication (Figure 4b). The data used to generate Figure 4 is additionally listed in Table 1 and Table 2. In these plots, the distinction between deposition categories is as follows. Molecular beam epitaxy (MBE), pulsed laser deposition (PLD), and (plasma enhanced) atomic layer deposition (PEALD) are considered *advanced vacuum* techniques, all sputtering variations as well as metal-organic chemical vapour deposition (MOCVD) are grouped as *vacuum* techniques, whereas methods such as spin coating, spray pyrolysis (50, 67) or inkjet printing (12, 68) are in the *solution* processing category.

The largest range of μ_{FE} is found within the advanced vacuum category with values between 6.5 for a $\text{In}_2\text{O}_3/\text{ZnO}$ HJ and $240 \text{ cm}^2/\text{Vs}$ for ZnO/ZnMgO (42, 69), with the latter value being close to Hall effect mobility of $250 \text{ cm}^2/\text{Vs}$ measured in a comparable ZnO/ZnMgO system at RT (23). This large variation can be explained mainly by two factors. First, the deposition temperature can have a major impact on the

resulting films, where typically higher temperatures lead to films with improved structural quality. Second, especially processes such as MBE require specialized substrates (*e.g.* sapphire) to reach the full potential of epitaxially grown materials (7, 23, 35, 40, 42). Common substrate materials, such as Si/SiO₂, glass etc., are far more commercially viable but do not yield the same quality films (15, 69-71). The range as well as maximum of reported mobility in the regular vacuum deposition and solution processing category is considerably smaller with values over 100 cm²/Vs (20, 72) and close to 50 cm²/Vs (43, 51, 54), respectively. The decline in performance is expected, as especially in the case of solution processing the films typically still contain a significant number of defects that hinder charge transport. Interestingly, there does not seem to be a strong correlation between the maximum temperature and the resulting mobilities for the HJ films in the vacuum and solution processing deposition categories.

Figure 4: Overview of mobility values achieved in heterojunction metal oxide structures, based on the values listed in Table 1 and Table 2. a) Mobility broken down by deposition categories, *i.e.* advanced vacuum deposition techniques (MBE, ALD, PLD), vacuum techniques (sputtering and MOCVD) and various solution processing techniques (spin coating, spray pyrolysis, inkjet). The color of each marker is indicative of the maximum substrate temperature during fabrication as shown in the color bar on the right. Within each category, markers are additionally distributed from left to right with increasing maximum temperature. b) Progression of mobility values with the year of publication. Here, the color of each marker denotes the deposition category for the MO semiconductors. The dashed lines are linear trend lines as guides for the eye. In both panels (a, b), square markers denote mobility values extracted via Hall effect measurements at RT whereas circular markers correspond to device mobility values extracted from TFTs.

As can be seen in Figure 4b, the initial studies on heterojunctions were started from 2003 to approx. 2008 (7, 23, 32, 35, 40, 42). Here, techniques such as MBE and PLD were used to grow high quality ZnO/ZnMgO HJ on sapphire substrates; an approach that lead to the highest reported mobility values. From 2008 onwards, it was attempted to reproduce the benefits of HJ films by more scalable and commercially viable vacuum deposition techniques, predominantly by sputtering on more common substrates (9, 17-21, 24, 28, 45, 47, 49, 58-61, 65, 66, 72-74). As a result, exceptionally high mobility values over 80 cm²/Vs were achieved in the ZnO/ZnMgO (47) system but also for ITO/GIZO (72) and ATZO/ATZO (20) (front and back channel processed with different oxygen pressure). Similarly, for solution processing techniques, research into HJMO TFTs was intensified from 2011 onwards, using spin coating in the vast majority of cases (8, 10, 11, 13, 14, 16, 25, 43, 44, 51-57, 62-64, 75, 76), but also including spray pyrolysis (50, 67) and inkjet printing (12, 68). This increasing shift in research efforts towards large scale and higher throughput fabrication techniques versus the ultrahigh quality epitaxial MO HJs of the initial phase can be seen as the reason why the overall average mobility actually appears to trend downwards over the years. However, if the advanced vacuum category is further divided into periods of epitaxial (before 2009) and polycrystalline growth (after 2009), within all categories the mobility trend-line is actually increasing over time (see dashed lines in Figure 4b).

Table 1: Overview of selected publications on heterojunction metal oxide films and TFTs processed by vacuum techniques. Entries with the added asterisk () in the mobility field denote Hall mobilities μ_H rather than TFT device mobilities μ_{FE} .*

Year	Heterojunction Structure	Deposition Method	Mobility (cm ² /Vs)	Dielectric	Maximum Temperature (°C)	Reference
		Deposition Category: Advanced Vacuum				
2003	ZnMnO:Al / ZnO	PLD	130 (*)	n/a	700	(32)
2004	ZnMgO/ ZnO / ZnMgO	MBE	170	n/a	750	(35)
2005	ZnMgO/ ZnO / ZnMgO	MBE	140	MgO	750	(40)
2007	ZnO / ZnMgO	MBE	240 170	HfO ₂ Al ₂ O ₃	500	(42)
2007	ZnO / ZnMgO	MBE	250 (*)	n/a	650	(23)
2011	ZnO / Al ₂ O ₃ / ZnO	PEALD	2.8	Al ₂ O ₃	200	(26)
2013	Al ₂ O ₃ /ZnO superlattice	ALD	27.8	SiO ₂	200	(15)
2015	ZnO / SnO multilayers	PLD	43	Al ₂ O ₃	350	(70)
2015	ZnO / HfO ₂	ALD	13	HfO ₂	160	(27)

2018	$\text{In}_2\text{O}_3 / \text{ZnO}$ supercycles	ALD	9.3	SiO_2	200	(71)
2018	$\text{In}_2\text{O}_3/\text{ZnO}$ supercycles	ALD	6.5	SiO_2	300	(69)
2019	$\text{In}_2\text{O}_3 / \text{ZnO}$ supercycles	PEALD	30.3	Al_2O_3	250	(77)
2019	$\text{In}_2\text{O}_3 / \text{ZnO} / \text{Ga}_2\text{O}_3$ supercycles	PEALD	74	SiO_2	450	(78)
		Deposition Category: Vacuum				
2008	IZO / GIZO or ITO / GIZO	Sputtering	51.3 104	SiO_2	300	(72)
2010	$\text{MgZnO} / \text{ZnO}$	MOCVD	9.1	Si_3N_4	450	(48)
2010	$\text{ZnO} / \text{MgZnO}$	Sputtering	39 (*)	n/a	600	(79)
2010	HIZO / HIZO Hf content varied	Sputtering	15	$\text{Si}_3\text{N}_4 / \text{SiO}_2$	370	(17)
2011	GZO / IGZO	Sputtering	10	SiO_2	200	(73)
2011	IGZO / IGZO:N	Sputtering	7.3	SiO_2	350	(18)
2011	ITO / ZTO	Sputtering	43.2	SiO_2	500	(58)
2011	IGZO / ITO	Sputtering	20	SiO_2	80	(49)
2012	IZO / HIZO	Sputtering	48	$\text{Si}_3\text{N}_4 / \text{SiO}_2$	350	(60)
2012	$\text{ZnO} / \text{MgZnO}$	Sputtering	84.2	Al_2O_3	600	(47)
2012	IZO / HIZO	Sputtering	41.4	SiO_2	300	(59)
2012	IZO / IGZO	Sputtering	47.7	SiO_2	250	(61)
2013	$\text{SnO} / \text{Cu}_2\text{O}$	Sputtering	0.66	Al_2TiO_5	190	(66)
2013	IGZO (O poor) / IGZO (O rich)	Sputtering	3.65	SiO_2	350	(19)
2014	IGZO / IGZO:Ti	Sputtering	63	HfO_2	300	(74)
2014	IGZO / IZO	IGZO: Spin coating IZO: Sputtering	46.7	Al_2O_3	430	(80)
2014	$\text{In}_2\text{O}_3 / \text{InZnO}$	Sputtering	37.9	ZrO_2	25	(9)
2014	IZO / ZTO	Sputtering	32.3	SiO_2	500	(65)
2016	IGZO:H / IGZO	Sputtering	55.3	SiO_2	250	(21)
2016	ATZO / ATZO varied O pressure	Sputtering	108	SiO_2	150	(20)
2017	$\text{In}_2\text{O}_3 / \text{Al}_2\text{O}_3$	Sputtering	20.5 (*)	n/a	200	(28)
2019	IGZO / In_2O_3	RF sputtering	64.4 67.5 79.1	SiO_2 HfO_2 Si_3N_4	25	(24)

Table 2: Overview of selected publications on heterojunction metal oxide TFTs processed by solution processing techniques. Entries with the added asterisk (*) in the temperature field were treated by photonic curing and the stated values are the approximate substrate temperatures during the process, based on simulations.

Year	Heterojunction Structure	Deposition Method	Mobility (cm^2/Vs)	Dielectric	Maximum Temperature ($^{\circ}\text{C}$)	Reference
2011	IZO / AIZO	Spin coating	1.57	Tetraethyl-orthosilicate	350	(62)
2012	IZO / AIZO	Spin coating	5.62	SiO_2	350	(63)
2013	IGZO / ZTO	Spin coating	2.09	SiO_2	450	(10)
2013	IGZO / IGZO / IGZO	Spin coating	2.4	SiO_2	450	(13)
2013	In_2O_3 / IGO	Spin coating	2.6	SiO_2	250	(8)
2014	IGZO / ZTO	Spin coating	1.97	SiO_2	450	(11)
2014	IZTO / IGZO	Spin coating	22.16	SiO_2	450	(57)

2015	$\text{In}_2\text{O}_3 / \text{Ga}_2\text{O}_3 / \text{ZnO} / \text{Ga}_2\text{O}_3 / \text{In}_2\text{O}_3$	Spin coating	30 40	SiO_2 $\text{Al}_2\text{O}_3/\text{ZrO}_2$	200	(14)
2015	$\text{ZTO} / \text{In}_2\text{O}_3$	Inkjet	8.6	SiO_2	400	(12)
2016	$\text{SnO}_2 / \text{ZnO}$	Spin coating	15.4	SiO_2	300	(64)
2017	$\text{ZTO} / \text{Al}_2\text{O}_3$	Spin coating	3.08	SiO_2	400	(25)
2017	$\text{In}_2\text{O}_3 / \text{ZnO}$	Spray pyrolysis and spin coating	45	SiO_2	250	(50)
2017	$\text{In}_2\text{O}_3 / \text{Li: ZnO}$	Spin coating	11.4	SiO_2	200	(53)
2017	$\text{In}_2\text{O}_3 / \text{GaOx}$	Spin coating	11	SiO_2	50	(75)
2017	$\text{In}_2\text{O}_3 / \text{ZnO}$	Spin coating	47.9	SiO_2	400	(51)
2017	$\text{In}_2\text{O}_3 / \text{ZnO}$	Spin coating	36	$\text{Al}_2\text{O}_3 / \text{ZrO}_2$	250 (*)	(52)
2017	$\text{ZnO} / \text{Ga}_2\text{O}_3$	Spray pyrolysis	1	SiO_2	233	(67)
2018	$\text{ITZO} / \text{IGZO (corrugated structure)}$	Spin coating	51	Al_2O_3	450	(43)
2018	$\text{ZnO} / \text{In}_2\text{O}_3$	Spin coating	0.5	SiO_2	350	(76)
2019	$\text{In}_2\text{O}_3 / \text{In}_2\text{O}_3:\text{PEI}$	Spin coating	10.05 24.8	SiO_2 ZrO_2	250 500	(56)
2019	$\text{In}_2\text{O}_3/\text{ZnO}:\text{Li}/\text{In}_2\text{O}_3/\text{ZnO}:\text{Li}$	Spin coating	18 25	SiO_2 $\text{Al}_2\text{O}_3/\text{ZrO}_2$	200	(16)
2019	$\text{In}_2\text{O}_3 / \text{ZnO NP:Al} / \text{PS} / \text{ZnO}$	Spin coating	50.7	SiO_2	200	(54)
2019	$\text{In}_2\text{O}_3 / \text{AlInO}$	Spin coating	40	SiO_2	300	(55)
2019	$\text{In}_2\text{O}_3 / \text{IGZO}$	Inkjet	14.5	SiO_2	400	(68)
2020	$\text{In}_2\text{O}_3 / \text{ZnO}$	Spin coating	19	$\text{Al}_2\text{O}_3/\text{ZrO}_2$	200 (*)	(44)

3.2 Distinct heterojunction layers versus mixed phase oxides

When investigating HJ stacks and their impact on device performance relative to single layers MO films, one important aspect is to verify whether any improvements are indeed the results of creating distinct layers and the subsequent interface effects taking place or if the same could have been achieved simply by creating a single multicomponent mixed phase oxide instead. Such a reference test was carried out for example by Khim *et al.* who researched solution processed $\text{In}_2\text{O}_3/\text{ZnO}$ HJs (16). As shown in Figure 5a, it was found that the IZO based reference devices, created by mixing the respective In- and Zn-containing precursors, showed poor transport behavior with low on-currents on the order of 10^{-6} A and a high turn on voltage of 26 V. Contrary, the In_2O_3 /bilayer exhibited superior properties with increased on-current (10^{-3} A) and a turn on close to 0 V. It was argued that the 200 °C annealing temperature did not supply enough thermal energy for a complete IZO conversion, whereas electron confinement at the successfully formed $\text{In}_2\text{O}_3/\text{ZnO}$ HJ was the reason for superior properties in the bilayer TFT. For a different material

system of solution processed SnO₂/ZnO, Nam *et al.* came to similar findings where TFTs based on control ZTO films showed inferior performance compared to the SnO₂/ZnO counterparts when annealed at a temperature of 300 °C (64).

3.3 Influence of layer thicknesses

The layer thicknesses of the individual metal oxides that form the heterojunction in the active TFT channel have a profound impact on the overall device characteristics. For example, the effect of the front channel thickness in solution processed IZO/AIZO HJs TFTs was investigated by Jeong *et al.* (63). As seen in Figure 5c, the on/off ratio decreases with increasing IZO front channel thickness between 8 and 20 nm. This is the result of increasing overall N_e for higher IZO thicknesses, which also lead to a negative V_{th} shift. The same trend was reported by Liu *et al.* for sputtered In₂O₃/IZO HJ TFTs where μ_{FE} steadily increases with higher In₂O₃ thickness, albeit at the cost of an increasing subthreshold swing (SS), decreasing on/off ratio and negative V_{th} shift (9). Consequently, a balanced compromise between high electron mobility and on-currents on one side and high on/off ratio, low SS and V_{th} close to 0 V on the other side, has to be made. While Lee *et al.* found very similar properties for increasing ITZO film thicknesses in their ITZO/IGZO HJ devices, they made use of these findings by changing up the channel configuration (43). They created corrugated bilayer films with stripes of thin ITZO/IGZO and thick ITZO/IGZO going perpendicular to the source-drain channel orientation. This procedure allowed for devices with high on-currents and mobility $> 50 \text{ cm}^2/\text{Vs}$ while simultaneously being able to maintain low off-currents and an on/off ratio $> 10^8$.

An investigation by Lee and Choi about inkjet printed ZTO/In₂O₃ bilayer HJ TFTs examined how the increase in In₂O₃ back channel thickness impacts the device performance (12). They found an initial improvement in μ_{FE} , on/off ratio, SS and V_{th} for thinner In₂O₃ films, which was attributed to the filling of

pores in the ZTO front channel as well as a decrease in roughness. Increasing the back channel further, however, saw a deterioration of all quantities due to the introduction of increased contact resistance, trap density and elevated roughness.

In a study on PLD deposited multilayer stacks consisting of repeated ZnO/SnO₂ units, the thickness of the individual layers was simultaneously varied between 0.5 and 3 nm while keeping the total channel thickness to 30 nm (70). The resulting transistors were found to exhibit a maximum μ_{FE} of 43 cm²/Vs for a thickness of 1.5 nm for both sub-units. Further increase in thickness only lead to a decline in performance. It was argued that high mobility electron channels formed at the ZnO/SnO₂ interfaces and that for higher sub-unit thicknesses the number of available interfaces decreased, thus reducing μ_{FE} in the device.

Krausmann *et al.* produced HJ TFTs based on multilayers with repeating In₂O₃/ZnO units via ALD and changed the thickness ratio between In₂O₃ and ZnO in the repeat units (71). They found that higher amounts of defects are introduced by a higher thickness of the ZnO layers, while opposite to that thicker In₂O₃ layers resulted in higher mobilities up to 12.2 cm²/Vs. However, other device parameters (on/off, SS, V_{th}) began to deteriorate for the thickest In₂O₃ sub-units and a more balanced optimum performance was determined for a medium size In₂O₃ thickness. In addition, the study revealed a gradual change in the transport properties from an initially trap-limited conduction for a low In₂O₃/ZnO thickness ratio towards a percolation conduction for thicker In₂O₃ layers in the repeating units. This also manifested in nearly temperature independent μ_{FE} as opposed to a temperature activated behavior with for devices with the lowest In₂O₃/ZnO ratios. For the same material system, with stacks of alternating In₂O₃ and ZnO processed by ALD, Sanctis *et al.* varied the total number of stacks between 3 and 6 (69). With higher number of stacks an increase in channel conductivity (reduced on/off ratio, negative V_{th} shift) was observed and attributed to deep-donor like states related to oxygen vacancies. An optimum number of 4 stacks was suggested based on the tradeoff between mobility, on/off ratio, subthreshold swing SS and V_{th}.

3.4 Influence of stacking sequence

In many bilayer HJ TFTs the more conductive MO layer is typically employed as the front channel material in direct contact with the dielectric. In this position it can serve as charge transport layer with a high N_e that also can assist in trap passivation at the semiconductor/dielectric interface (8, 9, 21, 71). This was evaluated and confirmed by Sanctis *et al.* who fabricated multilayer stacks of repeated In_2O_3 and ZnO layers via ALD (69). They tested the impact of starting the HJ sequence either with a layer of ZnO or In_2O_3 . For ZnO as the starting material, the multilayer TFTs yielded a low mobility of $1 \text{ cm}^2/\text{Vs}$, an on/off ratio of 5×10^6 and SS of 1.85 V/dec . All these quantities were found to be significantly improved when the sequence was reversed and In_2O_3 was the starting material, giving rise to a mobility, on/off ratio and SS of $6.5 \text{ cm}^2/\text{Vs}$, 5×10^7 , and 0.7 V/dec , respectively.

Another study on the influence of the stacking sequence for the same $\text{In}_2\text{O}_3/\text{ZnO}$ material system was carried out by Khim *et al.* (16). Here, film stacks of up to four sequentially solution deposited single layers were formed, alternating between In_2O_3 and ZnO in consecutive deposition steps. Film as well as device properties were evaluated at each stage. Additionally, the films stack fabrication was either started by the deposition of In_2O_3 or ZnO as the initial layer. As shown in Figure 5d and e, the progression of device characteristics with each new layer was vastly different. For stacks beginning with In_2O_3 , the mobility increases with each new steps (from 1.6 to $11.4 \text{ cm}^2/\text{Vs}$), making bigger jumps after a new ZnO film was deposited. In contrast to that, the devices using ZnO as the starting layer were limited by the mobility of a single ZnO film ($1.4 \text{ cm}^2/\text{Vs}$) when ZnO is the topmost layer and μ_{FE} even dropped about 2 orders of magnitude when In_2O_3 is the topmost layer. Based on AFM investigations, it was determined that the interface roughness plays a crucial role, whereas the $\text{In}_2\text{O}_3/\text{ZnO}$ interface is favorable for charge transport but the $\text{ZnO}/\text{In}_2\text{O}_3$ interface is doing the opposite. Consequently, sequences starting with In_2O_3 inherently include more beneficial HJs that have an accumulative effect, while sequences starting with ZnO as the

initial material contain more unfavorable ZnO/ In_2O_3 junctions leading to more traps in the system and limited device performance.

Figure 5: a) Comparison between HJ TFT with an active channel comprised of solution processed $\text{In}_2\text{O}_3/\text{ZnO}$ and a device based on a single blend layer of IZO using the same precursor components. The distinct HJ device exhibits superior performance. Reproduced with permission. (16) Copyright 2019, John Wiley and Sons. b) Effect of MOD on the transfer characteristics in $\text{In}_2\text{O}_3/\text{ZnO}$ HJ TFTs via Li-doping of the ZnO layer. Reproduced with permission. (53) Copyright 2017, John Wiley and Sons. c) Evolution of transfer curves for TFTs based on solution processed IZO/ Al_2O_3 HJs with increasing IZO front channel thickness between 8 and 20 nm. © 2012 IEEE. Reprinted, with permission, from (63). d) and e) The effect of stacking sequence on μ_{FE} for In_2O_3 (I) and ZnO (Z) multilayer TFTs. Reproduced with permission. (16) Copyright 2019, John Wiley and Sons.

3.5 Modulation doping

Modulation doping (MOD) is a technique to improve the device performance wherein two separate semiconductors are brought into close contact, one of them is undoped (intrinsic) while the other one is intentionally doped. Given the correct relative energy levels, an electron flow (in the case of n-type doping) from the doped to the undoped layer is initiated. Because the ionized dopant sites are physically

separated from the undoped semiconductor the charge transport in the latter is not hampered by ionized impurity scattering, enabling higher carrier mobilities. Modulation doping finds application for example in GaAs/(Al,Ga)As high electron mobility transistors (29).

The introduction of MOD for MO HJ structures was simulated for the ZnO/ZnMgO system (81) and also experimentally verified for sputtered films, where the ZnMgO layer was doped with a fixed concentration of Al (47, 79). The addition of the doping layer lead to an increased sheet carrier density in addition to the polarization induced charge carries. When the Mg content in the doping layer was varied, the band gap increased with increasing Mg concentration and as a result the tendency of electron transfer to the 2DEG region increased, resulting in decreased sheet resistance and increased sheet carrier densities. According to their estimates, Chin *et al.* suggested that up to 61 % of the charge carriers in the heterostructure originated in the doping layer (79).

Regarding solution processed HJs, Khim *et al.* used Li-doped ZnO films to apply MOD in In₂O₃/ZnO bilayer TFTs (53). Increasing the Li concentration from 0 to 20 wt% (in the precursor solution) was found to move the ZnO Fermi level (E_F) closer to vacuum due to higher electron concentrations in the doped ZnO films. The resulting increased offset between the Fermi levels of In₂O₃ and ZnO:Li was suggested to facilitate electron transfer from ZnO:Li to In₂O₃. Modulation doping lead to significantly reduced sheet resistance of bilayer films and the μ_{FE} of HJ TFTs saw an improvement from 3.2 to 11.4 cm²/Vs for 0 to 20 wt% Li doping. Figure 5b shows the evolution of transfer curves depending on the doping concentration. In a follow up study, MOD with Li was also used to boost the performance of MO multilayer TFTs based on a In₂O₃/ZnO/In₂O₃/ZnO channel architecture (16). Here, an increased mobility of 18 cm²/Vs and improved uniformity could be demonstrated using a SiO₂ dielectric as well as μ_{FE} up to 25 cm²/Vs using a high-k dielectric based on AlO_x/ZrO_x. These results are a good indicator for the improvements that can be achieved using MOD.

4 Improvements in bias stress stability for heterojunction TFTs

For most of the targeted applications of HJMO TFTs, each single TFT is but one device out of a large group, working together to fulfill a specific system function. One simple example is an individual transistor switching on (or off) a single pixel as part of a display that comprises many thousands of pixels. To assure the overall functionality is not compromised, it is therefore critical that each individual device stays within its operational parameters, even over prolonged times of activity and in different environments. Bias stress tests are one common way of testing a device's resistance against the application of constant biases over extended periods of time.

In a typical bias stress (BS) test, an initial transfer curve is recorded first, then a fixed set of drain voltage V_D and gate voltage V_G is applied over a long time frame, with intermittent transfer curve measurements at certain intervals. Depending on the polarity of V_G and ambient conditions, different types of bias stress tests are defined:

PBS: Positive bias stress, *i.e.* V_G is set at a positive value (9-13, 15, 16, 19, 21, 25, 54, 57, 64, 68).

NBS: Negative bias stress, *i.e.* V_G is set at a negative value (11, 19, 21, 25, 61, 68).

NBIS/PBIS: Negative/positive bias stress under constant illumination (11, 25, 26, 61).

NBTS/PBTS: Negative/positive bias stress including thermal stress at elevated temperatures (25, 64).

As it is the most commonly performed test, we will focus on PBS in the following examination. While other values, such as the subthreshold swing SS or mobility μ_{FE} can be taken into consideration, most often the shift in threshold voltage V_{th} over time is used to assess the severity of bias stress on the devices. A bias stress comparison between different literature reports is challenging as the magnitude of V_{th} and its shift over time is influenced not only by the active semiconductor channel composition but also heavily by the choice of gate dielectric. Additionally, there is no clear and commonly agreed upon, framework regarding

what voltage conditions should be applied during the testing and how long a bias stress test should be run for. While most investigations were carried out for durations below 3 h (9-13, 15, 19, 21, 25, 57, 68), only a few reports feature measurements up to 9-10 h (16, 64) or even up to 24 h (54). In an effort to facilitate comparisons between bias stress tests, Lin *et al.* (54) proposed to calculate the electric flux density D and electron trap density N_{tr} according to the equations below:

$$D = C_i V_{G,PBS} \quad (1)$$

$$N_{tr} = \frac{C_i |\Delta V_{th}|}{e} \quad (2)$$

with e being the electron charge. These quantities incorporate device and measurement parameters, namely the geometric gate capacitance C_i , the applied gate voltage $V_{G,PBS}$ as well as the shift in threshold voltages (ΔV_{th}) between the initial and final transfer curve during the bias stress procedure. They are therefore more meaningful to compare than the raw measure of ΔV_{th} . Note, however, that this approach is not taking into account the value of V_D during the bias stress procedure. Higher drain voltages can reduce the ΔV_{th} through a field-assisted de-trapping mechanism (Poole-Frenkel effect) (54).

The threshold voltage shift during PBS application is typically an effect of various sources of trap states, which capture electrons from the channel when they are filled. Consequently, as the stress time increases, fewer and fewer electrons can actively contribute to the drain current, unless the gate bias is increased as well in order to compensate for this effect. As a result, the transfer curve and V_{th} are shifting to more positive gate voltages in the course of a PBS experiment. Trap states can manifest as the result of various defects, such as *e.g.* oxygen vacancies V_O , imperfect metal-oxide-metal (M-O-M) network formation or impurities (25). Another source of trap sites can be seen in the top-surface of the MO channel, which can interact with ambient gases in the environment. Specifically, the presence of oxygen during PBS causes

adsorption on the channel surface, leading to the formation of O_2^- species as electrons are captured from the channel.

To improve the bias stress stability of a device, each of these possible trap sites should be addressed. Inherent defects caused by *e.g.* incomplete M-O-M formation, remaining film porosity or impurities are often a result simply of the deposition method, where low-temperature solution processed films typically show the highest trap numbers. This might be mitigated to some degree, either by novel approaches in precursor design (82, 83), higher process temperatures or new post-deposition treatments (44, 52), or simply changing to more advanced vacuum deposition techniques. However, for various reasons this is not always feasible, *e.g.* regarding process constraints (temperature sensitive substrates, large scale manufacturing), or cost considerations. Furthermore, as a means to reduce the number of V_O , it is a common strategy to introduce elements with a strong affinity towards oxygen into the MO matrix, such as Ga, Hf, or Al. Together with the desired V_O reduction, however, this often has the unwanted side effect to lower the charge carrier mobility (13, 21). It is therefore challenging to simultaneously realize high μ_{eff} and a good bias stress stability (low trap density) within a single layer MO channel.

This is where heterojunction transistors come into play because they allow to tackle both targets at the same time. A common approach is to combine a front channel material with a high mobility μ_{FE} and carrier density N_e together with a back channel with of lower mobility but increased stability. The higher carrier density helps to passivate large number of (interface) traps and the backchannel additionally adds separation between the charge transporting channel near the dielectric and any gas molecule interactions at the top surface. Additionally, the back channel may be processed in a way to minimize the likelihood of oxygen adsorption, *e.g.* by creating an oxygen rich material during the deposition (19).

Bias stress investigations were carried out in such kind of HJ structures with different material combinations as front and back channel. For example, Abliz *et al.* used a homojunction of IGZO/IGZO in

which the front channel underwent a hydrogen plasma to increase N_e (21). Similarly, Zhan *et al.* varied the carrier concentrations in both components of an IGZO / IGZO bilayer by adjusting the oxygen pressure during deposition via RF sputtering, thus creating an oxygen-poor but conductive front channel and an oxygen-rich IGZO back channel (19). Kim *et al.* used solution processed IGZO film stacks with an increased Ga content for the top-most layer (13). Other combinations rely on a front channel of In_2O_3 combined with IZO (9), ZnO (16), or IGZO (68) as the back channel. Exceptionally, In_2O_3 was only found as the back channel material in the study by Lee *et al.* (12). In a recent study, Lin *et al.* employed a solution processed $\text{In}_2\text{O}_3/\text{ZnO}$ bilayer and improved upon the trap passivation by inserting both ZnO nanoparticles (NPs) as well as a thin, UV/ozone treated layer of polystyrene (PS) in between the front and back channel (54). This hybrid organic-metal oxide multilayer approach lead to exceptional improvements in PBS stability.

As non In-containing HJ channel materials for bias stress tests, the combinations of SnO_2/ZnO (64) or $\text{ZTO}/\text{Al}_2\text{O}_3$ (25) were investigated. Al_2O_3 was additionally studied by Ahn *et al.* as part of their $\text{ZnO}/\text{Al}_2\text{O}_3$ superlattices processed by ALD (15).

For all of the studies and HJ material combinations mentioned above, the effect was an increased resistance against bias stress. An overview comparing the results of various reports for PBS measurements of HJ TFTs is given in Figure 6 and Table 3. Figure 6a shows the trap density (as measure for the shift in transfer characteristics throughout the BS measurements) depending on the electric flux density (as a quantity describing the magnitude of applied stress). Several aspects are immediately apparent. First of all, there are two distinct groupings, one at lower values of D between 0.23 to $1.4 \times 10^{-6} \text{ C cm}^{-2}$ and a single data point at around $2.3 \times 10^{-6} \text{ C cm}^{-2}$. This reflects the fact that to date, most BS investigations on HJ-MO TFTs were carried out using bottom-gate top contact device architecture with a Si wafer as combined substrate and gate electrode, with a SiO_2 layer atop acting as the gate dielectric. The SiO_2 thickness ranges from 100 to 300 nm and the $V_{G,\text{PBS}}$ between 20 and 40 V, leading to the variation seen in the electric flux. On the contrary, the investigation by Liu *et al.* used a $\text{In}_2\text{O}_3/\text{InZnO}$ HJ on top of solution processed ZrO_2

dielectric with a high C_i of 775 nF cm^{-2} which results in a high electric flux density over $2 \times 10^{-6} \text{ C cm}^{-2}$ even at low $V_{G,\text{PBS}}$ of 3 V (9). While there are obviously differences between the HJ systems, as a rough trend, the resulting trap density increases with increasing electric flux, with the highest value being $1.9 \times 10^{12} \text{ cm}^{-2}$. This intuitively makes sense as a more pronounced ΔV_{th} is expected when a higher gate bias is used during PBS. Finally, the color-coding in Figure 6a refers to the total stress time, from which it can be seen that most PBS investigations to date are kept below 3 h while only a few are carried out for times >9 h (16, 64), with the longest one by Lin *et al.* reaching up to 24 h (54).

Figure 6: Overview of PBS literature results for metal oxide heterojunction TFTs. a) Trap density shifts for HJ TFTs over electric flux density applied during PBS application. The color coding of the marker indicates the total stress time up to 24 h. b) Trap density shifts after PBS application for HJ TFTs (blue) and their single layer counterparts (red). In both panels, the shape of the marker indicates the deposition category, broken down to advanced and regular vacuum techniques as well as solution processing, as described earlier.

As pointed out before, one of the benefits of using a HJ approach is in the improved bias stress stability. This is visualized in Figure 6b where the resulting values of trap density for a HJ TFTs are shown in comparison to a reference device based on a single layer channel material. The single layer films span over a wide range of trap densities between 0.24 and $3.7 \times 10^{12} \text{ cm}^{-2}$. Accordingly, the changes in trap density when going towards a HJ device vary over a similar range. Although not a clear correlation, the highest margins seem to be possible when the initial single layer TFT shows increased levels of N_{tr} , whereas the

room for improvements is more limited with a low starting point. When comparing the different deposition methods (see marker shapes in Figure 6b), the lowest trap density is achieved using atomic layer deposition (15) while higher values are the result of less sophisticated methods such as sputtering techniques or via solution based processes. Especially solution-processed layers are often inherently more prone to defects due to *e.g.* non-fully converted precursors material, residual contaminants, or pinholes. As a result, those kinds of films can potential gain the most benefits of forming a heterojunction through the deposition of a second layer, which allows an effective defect compensation and thus more stable devices under bias stress.

Table 3: Overview of PBS investigations on metal oxide HJ structures

Year	Heterojunction Structure	Deposition Method	Dielectric	C_i (nF/cm ²)	$V_{G,PBS}$ (V)	ΔV_{th} (V)	Stress Time (s)	D (10 ⁻⁶ C/cm ²)	N_{tr} (10 ¹² /cm ²)	Reference
2011	IGZO / IGZO:N	Sputtering	SiO ₂	34.5	10	3.7	10,000	0.34	0.8	(18)
2013	Al ₂ O ₃ /ZnO superlattice	ALD	SiO ₂	17.3	30	0.5	7,200	0.52	0.05	(15)
2013	IGZO / ZTO	Spin coating	SiO ₂	28.8	20	0.6	1,000	0.58	0.11	(10)
2013	IGZO / IGZO / IGZO	Spin coating	SiO ₂	28.8	20	4.2	1,000	0.58	0.75	(13)
2013	IGZO (O poor) / IGZO (O rich)	Sputtering	SiO ₂	34.5	20	3.08	1,500	0.69	0.66	(19)
2014	IGZO / ZTO	Spin coating	SiO ₂	28.8	20	1.51	10,000	0.58	0.27	(11)
2014	ITZO / IGZO	Spin coating	SiO ₂	34.5	20	5.01	10,000	0.69	1.08	(57)
2014	In ₂ O ₃ / InZnO	Sputtering	ZrOx	775.0	3	0.4	9,000	2.33	1.93	(9)
2015	ZTO / In ₂ O ₃	Inkjet	SiO ₂	11.5	20	9.45	3,600	0.23	0.68	(12)
2016	IGZO:H / IGZO	Sputtering	SiO ₂	34.5	20	1.1	3,600	0.69	0.24	(21)
2016	SnO ₂ / ZnO	Spin coating	SiO ₂	34.5	20	1.98	36,000	0.69	0.43	(64)
2017	ZTO / Al ₂ O ₃	Spin coating	SiO ₂	17.3	20	2.4	4,200	0.35	0.26	(25)
2019	In ₂ O ₃ / ZnO NP:Al / PS / ZnO In ₂ O ₃ / ZnO NP / PS / ZnO	Spin coating	SiO ₂	34.5	30	2.25 1.1	86,400	1.04	0.48 0.24	(54)
2019	In ₂ O ₃ / IGZO	Inkjet	SiO ₂	11.5	20	1.8	3,600	0.23	0.13	(68)
2019	In ₂ O ₃ / ZnO	Spin coating	SiO ₂	34.5	40	4.05	32,400	1.38	0.87	(16)

5 Conclusions

The exploration of metal oxide heterojunctions and their implementations in thin-film transistors is an exciting field of current research, ranging from basic research on 2DEG formation and transport physics on one side, to simultaneous improvements of mobility and bias stress stability for potential commercial applications in various display technologies, on the other. Especially, for deposition techniques that are geared more towards low-cost and high throughput fabrication on large area substrates, the formation of HJ structures is an approach that offers many benefits. Due to the multitude of possible combinations of HJ bilayers and even more so in connection with multilayer junctions, in terms of their composition and stacking sequences, many avenues are yet to be investigated. Advancements in interface and band gap engineering of metal oxide materials for HJs promise superior device properties that are out of reach for single metal oxide materials.

In addition to the improvements of current metal oxide HJ concepts, the transfer of knowledge to further domains in the future is envisioned. Especially with regards to p-type oxides, the integration of organic materials into organic/inorganic hybrids or even considering sequences fully based on organic films, it will be exciting to see how far the idea of heterojunctions can be extended. As a whole, an increased usage of HJ structures is expected to open new windows of opportunities for next generation (flexible) electronic applications.

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