Epitaxial Growth of $\kappa-(Al_{x}Ga_{1-x})_2O_3$ Layers and Superlattice Heterostructures up to $x = 0.48$ on Highly Conductive Al-Doped ZnO Thin-Film Templates by Pulsed Laser Deposition

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(Al$_{x}$Ga$_{1-x}$)$_2$O$_3$ thin-film layers in the metastable orthorhombic $\kappa$-modification are deposited heteroepitaxially on unintentionally doped ZnO buffer layers on heavily Al-doped and highly conductive ZnO back contact layers by pulsed laser deposition. The back contact layers potentially serve as electrodes for ferroelectric hysteresis measurements and improve the performance of device applications such as Schottky barrier diodes or quantum-well infrared photodetectors (QWIPs). The alloy layers follow a linear evolution with Al content in agreement with Vegard’s law. Smooth surface morphologies are verified in atomic force microscopy images. Finally, a 15-layer-pair $\kappa-(Al_{0.27}Ga_{0.73})_2O_3/\kappa-Ga_2O_3$ quantum-well superlattice (SL) structure is similarly deposited on the ZnO:Al back contact layer and shows sharp SL fringes up to the fifth order in XRD measurements confirming excellent crystal quality and abrupt interfaces in the heterostructure. The results render ZnO:Al as a promising back contact layer for QWIP applications and the determination of electrical polarizations of $\kappa$-phase alloy layers.

1. Introduction

During the past decade, the wide-bandgap semiconductor Ga$_2$O$_3$ has found tremendously growing interest. The potential use for power-device applications of the material based on its large predicted electrical breakdown field of about 8 MV cm$^{-1}$ for the monoclinic $\beta$-modification\cite{1} initially triggered these research efforts. The possibility of the homoeptaxial growth of devices using commercially available single-crystal $\beta$-Ga$_2$O$_3$ wafers up to 4” diameter intensified the interest even further. This is substantiated by a tremendous amount of research papers on different phases of Ga$_2$O$_3$ that emerged into a large number of recent reviews.\cite{1,2,3} Lately, metastable polymorphs of Ga$_2$O$_3$ came into the focus of research. Especially the orthorhombic $\kappa$-modification (also referred to as $\kappa$-Ga$_2$O$_3$) is of large interest as it is the only known polymorph predicted to exhibit a spontaneous electrical polarization of $\approx 23 \mu$C cm$^{-2}$ along its $c$-direction.\cite{4,5,6}

For comparison, this value is three times higher than the one of AlN and even one order of magnitude larger than for GaN.\cite{6,7,8} Similar to GaN and its alloy systems, this property can be utilized for polarization doping, where polarization discontinuities at heterointerfaces, e.g., to the $\kappa-(Al_{x}Ga_{1-x})_2O_3$ or $\kappa-(In_{x}Ga_{1-x})_2O_3$ alloy system, are used that result in polarization charges. When these polarization charges are compensated by free electrons, a 2D electron gas (2DEG) can form, which can be utilized in devices such as high-electron-mobility transistors (HEMTs) or quantum-well infrared photodetectors (QWIPs) without extrinsic doping. Research on such devices in the GaN, AlGaN, and InGaN systems has been conducted for years. Due to the larger expected polarization differences for the Al- and In-alloy systems of $\kappa-(In_{x}Ga_{1-x})_2O_3$, even higher 2DEG densities can be expected. Recent theoretical calculations substantiate these expectations.\cite{9} Further, the larger bandgap of $\kappa$-Ga$_2$O$_3$ of about 4.9 eV\cite{10,11,12} compared with GaN\cite{13} ($E_g \approx 3.4$ eV) can push the transparency of the devices toward...
solar-blind regions enabling infrared detection for extraterrestrial applications.

κ-Ga2O3 can be grown heteroepitaxially by several deposition methods, such as pulsed-laser deposition,[14,19–26] (PLD), halide vapor phase epitaxy,[18,27–29] (HVPE), metal-organic chemical vapor deposition,[17,30–35] (MOCVD), metal-organic vapor phase epitaxy,[36–39] (MOVPE), atomic layer deposition,[31] molecular beam epitaxy[40] (MBE), plasma-assisted molecular beam epitaxy[41] (PAMBE), and mist CVD.[15,16,42–49]

Electrochemically conducting thin films, which are a prerequisite for device applications, have been reported recently by Parisini et al.[37] Their thin films were grown by MOCVD and doped in situ (ex situ, by indiffusion) by silicon (tin). However, for deposition methods like PLD and MBE, tin typically does not act as an active donor, but it is necessary to stabilize the κ-phase.[14,19,20,23,40]

Research on the In as well as Al alloy systems of the κ-phase is still relatively scarce, as up to now only reports on PLD[21–25,50,51] as well as mist CVD[46,48] growth are available. However, these alloys can be grown phase pure in a broad composition range[23–25,50,51] of $x_{\text{In}} \leq 0.35$ and $x_{\text{Al}} \leq 0.65$. The energetic position of the valence band maximum was further found to be independent of the alloy composition[51] giving rise to large possible conduction band offsets and widely tunable wavelength ranges in QW κ-Ga2O3 thin-layer superlattices for QWIP applications ($\Delta E_g \approx 2.5 \text{ eV}$ for highest Al and In contents).

Further, we recently reported on the coherent growth of high-quality κ-(Al$_{0.5}$Ga$_{0.48}$)$_2$O$_3$/κ-(In$_{0.35}$Ga$_{0.65}$)$_2$O$_3$ heterostructures for QWIP applications with different Al contents of $x = 0.48$ with the same high crystalline quality as for comparable layers grown directly on single-crystalline c-sapphire substrates. Epitaxial relationships of the κ-phase layers to the ZnO growth template as well as the evolution of in- and out-of-plane lattice constants with Al content will be determined and evaluated as well. The surface morphology of the layers will be investigated by atomic force microscopy (AFM) validating smooth surfacemorphologies independent on Al content. Finally, to confirm the potential of κ-ZnO as back contact layer and growth template in sophisticated QWIP applications, we will investigate a κ-(Al$_{0.5}$Ga$_{0.48}$)$_2$O$_3$/κ-Ga$_2$O$_3$ QW SL heterostructure grown on a back contact layer and will prove that it shows the same crystalline properties as SL heterostructures deposited on single-crystalline c-sapphire substrates as well. The results suggest κ-ZnO as excellent growth template and back contact layer for applications in device structures based on κ-Ga$_2$O$_3$ and the remaining exact determination of electrical polarizations aiding the simulation of these devices.

2. Results and Discussion

2.1. Structural Properties and Crystalline Quality

We first investigate the structural properties of the κ-Ga$_2$O$_3$ thin-film samples on the ZnO:ZnO:Al template layers by means of XRD 2θ–ω scans as well as XRD rocking curves. Figure 1a shows typical XRD 2θ–ω scans of the samples with different Al contents of $x = 0$, 0.14, 0.28, and 0.48. The prominent reflection at 2θ ≈ 34.5° belongs to the (00.2) plane of ZnO confirming a (00.1) oriented growth of the template layers on the a-sapphire substrate as expected from literature.[60]

Apart from the Al$_2$O$_3$ substrate and ZnO template reflections,
only reflections indicating a growth of the $(Al,Ga)_{1-x}O_3$ layers in the $κ$-modification with a (001) orientation can be identified in the whole measurement range of $10^\circ$–$130^\circ$. Namely, these are the (002), (004), (006), and (008) reflections marked with an orange asterisk in Figure 1a, where only the first three are shown. For $x=0.48$, an additional binary $κ-Ga_2O_3$ buffer layer was needed to stabilize the $κ$-phase for Al contents larger than 40 at%,

$$\frac{a_{Al}}{a_{Ga}}=\frac{\sqrt{3}}{2}$$

such that reflections of the buffer layer are present in the scan as well, marked with a black dashed line. The $κ$-phase (006) reflections occur at slightly larger positions than $\bar{κ}-Al_{2}O_{3}$ (002), (004), and (006) reflections. For $x=0.28$, an additional $κ-Ga_2O_3$ buffer layer had to be introduced prior to deposition of the alloy layer such that reflections of binary $κ-Ga_2O_3$ are present in the diffractogram. The angular positions are given relative to the $Al_{2}O_{3}$ (11.0) substrate reflection also present in the scans. The angular positions are given relative to the $Al_{2}O_{3}$ (11.0) substrate reflection also present in the scans.

**Figure 1.** a) Typical XRD 2θ–ω scans of a series of $κ-(Al,Ga)_{1-x}O_3$ thin-film layers grown on $ZnO/ZnO:Al$ templates on a-sapphire substrates. Al content $x$ as indicated. Peaks marked with orange asterisks belong to the (002), (004), (006), (008), and (0010) reflections discussed later. b) Out-of-plane $c$-lattice constant of the $κ-(Al,Ga)_{1-x}O_3$ alloy layers as a function of $x$. The red dashed line is a linear fit to the data. c) XRD rocking curves of the $κ-(Al,Ga)_{1-x}O_3$ thin-film layers shown in (a) recorded around the symmetric $κ-(Al,Ga)_{1-x}O_3$ (004) reflection. The angular positions are given relative to the $Al_{2}O_{3}$ (11.0) substrate reflection also present in the scans. d) FWHM of the $κ-(Al,Ga)_{1-x}O_3$ (004) reflections as a function of $x$ extracted from the rocking curves and the XRD 2θ–ω scans shown as black squares and blue circles, respectively.

oriented growth is typical for the $κ$-phase apparently regardless of substrate orientation and material, as it is the only orientation reported in literature so far. The $κ-(Al,Ga)_{1-x}O_3$ (00n) reflections show low broadening with prominent $K_{\alpha1}/K_{\alpha2}$ splitting starting at the (004) reflection and shift to larger angular positions with increasing $x$. The latter is expected by reports on $κ-(Al,Ga)_{1-x}O_3$ growth on c-sapphire in literature

$$\frac{a_{Al}}{a_{Ga}}=\frac{\sqrt{3}}{2}$$

and is caused by decreasing lattice constants due to the smaller ionic radius of Al in comparison with Ga. The out-of-plane $c$-lattice constant of all samples was determined by extrapolation of the (001) lattice plane distances calculated from the angular positions of all (00n) reflections to $θ=90^\circ$ using a regression function to minimize the goniometer error, as described in previous studies.

| $x$ | $c$-lattice constant (Å) |
|-----|-------------------------|
| 0.28 | 9.30 |
| 0.48 | 9.10 |

The $c$-lattice constant as a function of $x$ is shown in Figure 1b. The value as determined from the positions of the barrier layer reflections for the Si sample is already included here, the corresponding XRD 2θ–ω scans are shown in Figure 5a.

The $c$-lattice constants are linearly decreasing with $x$ as expected from literature. A linear fit to the data gives the relation

$$c(x) = c_0 - kx$$

where $c_0$ is the value at $x=0$ and $k$ is the slope.
This is, within the experimental error, the same relation as obtained by us before for $\kappa\langle\text{Al}_x\text{Ga}_{1-x}\rangle\text{O}_3$ layers on c-sapphire substrates.\textsuperscript{24,25}

To further evaluate the crystalline quality of the samples, XRD rocking curves of the (004) reflection of the samples were recorded. These are shown in Figure 1c where $\omega$ is given relative to the position of the (11.0) reflection of the a-sapphire substrate to account for sample tilts. The $\kappa\langle\text{Al}_x\text{Ga}_{1-x}\rangle\text{O}_3$ (004) reflection shifts to larger angular positions with increasing $x$ as well, together with a slight increase in broadening of the reflections. To quantify this, we determined the broadening of the (004) reflections as full width at half maximum (FWHM) both from the rocking curves and from the $2\theta$–$\omega$ scans. These are shown in Figure 1d as a function of $x$. The broadening in the rocking curves is relatively large and increases from 0.65$^\circ$ up to 0.94$^\circ$ from $x = 0$ to $x = 0.28$. Then it slightly decreases again for $x = 0.44$, which is probably due to the binary $\kappa\text{Ga}_2\text{O}_3$ buffer layer, an effect that has been observed before for $\kappa\langle\text{Al}_x\text{Ga}_{1-x}\rangle\text{O}_3$ thin films by us\textsuperscript{50} and which might be caused by an increased crystalline quality of the alloy due to the buffer layer. The large broadenings are supposedly caused mainly by the nanocrystalline growth of the layers in three rotational domains, i.e., a low lateral X-ray coherence length, and not due to an increased mosaicity of the layers as discussed later and in previous studies.\textsuperscript{19,24,30} Further, they are similar to broadenings of $\kappa\text{Ga}_2\text{O}_3$ layers on c-sapphire substrates reported before.\textsuperscript{19} The broadening in the $2\theta$–$\omega$ scans is almost independent of $x$ in the range of 0.07 – 0.08$^\circ$, which is close to the resolution of the experimental setup ($\approx 0.05^\circ$). This is also similar to the broadenings of thin films on c-sapphire reported by us earlier.\textsuperscript{19} Therefore, the thin films can be grown on ZnO:Al back contact layers with similar high quality for all $x$ as for the growth on other substrates.\textsuperscript{19,24}

### 2.2. Epitaxial Relationships and Lattice Constants

The in-plane epitaxial relationships were investigated by XRD $\phi$ scans of skew-symmetric reflections of a c-sapphire substrate [[(11.3)], ZnO template [(10.1)], and $\kappa\langle\text{Al}_x\text{Ga}_{1-x}\rangle\text{O}_3$ layers [(131) and (122)]. Figure 2a shows typical $\phi$ scans for a binary $\kappa\text{Ga}_2\text{O}_3$ layer. The respective measurements for the other Al contents and the SL sample look similar and are shown in the Supporting Information (Figure S1, Supporting Information). The ZnO thin-film template grew without rotational domains as apparent from the six-fold occurrence of the (004) reflection. From the relative positions to the (11.0) reflections of the $\kappa\text{Ga}_2\text{O}_3$ layers, the epitaxial relationships can be given as ZnO [(11.0)]](00.1)Al$_2$O$_3$ and ZnO [00.1]](11.0)Al$_2$O$_3$, which agree with previous reports.\textsuperscript{60} The epitaxial growth of the $\kappa\text{Ga}_2\text{O}_3$ layer is apparent as well. The six-fold splitting of the (131) reflection as well as the occurrence and 12-fold splitting of the (122) reflection indicate both unambiguously the growth in the $\kappa$-phase and orthorhombic symmetry of the layer (in contrast to an also proposed hexagonal symmetry, for which the polymorph is often called $\kappa\text{Ga}_2\text{O}_3$) and a growth in three rotational domains twisted by 120$^\circ$ with respect to each other. A detailed discussion on this can be found in previous studies.\textsuperscript{19,50} From the position of the (131) reflection on the same angular position as the ZnO (10.1) reflection, the epitaxial relationships $\kappa\text{Ga}_2\text{O}_3$ [(100)](10.0)ZnO, $\kappa\text{Ga}_2\text{O}_3$ [(010)](12.0)ZnO, and $\kappa\text{Ga}_2\text{O}_3$ [(001)](010)ZnO can be deduced. The possible configuration of the three rotational domains of $\kappa\text{Ga}_2\text{O}_3$ on the (00.1) plane of ZnO is schematically depicted in Figure 2b. Interestingly, these in-plane epitaxial relationships correspond to a rotation of the $\kappa\text{Ga}_2\text{O}_3$ unit cell by

\begin{equation}
\text{c}(x)(\text{Å}) = (9.270 \pm 0.002) - (0.325 \pm 0.005)x
\end{equation}
30° with respect to the one for the growth on c-sapphire, where the [100] a-axis of κ-Ga2O3 is parallel to the m-plane facets of the sapphire. In this case, it is parallel to the a-plane facets of ZnO. This is possibly caused by the lattice mismatch, which is minimized for this configuration with −10.36% in a-direction and −10.75% in b-direction calculated using the unit cell values for ZnO from JCPDS card no. 79-2205 and for κ-Ga2O3 as given by Cora et al.\textsuperscript{[46]} In comparison with other substrates, this is quite large (e.g., ≈6% for c-sapphire\textsuperscript{[19,47]} and −(2−3)% for MgO (111)\textsuperscript{[19]}), but this does not seem to impair the crystalline quality of the layers significantly. This might be due to the fact that the alloy typically grows relaxed for the thicknesses investigated here,\textsuperscript{[24,26]} such that only the first few nm should be affected by the lattice mismatch or a thin interfacial layer in a different phase not detectable by XRD compensates the mismatch as was already observed before on other substrates.\textsuperscript{[16,62,63]}

However, the mismatch is still lower than for ITO as alternative back contact layer for κ-phase thin films (−15.9% in a and 12.7% in b-direction\textsuperscript{[16]}. Further, κ-phase layers on ITO typically show an inferior crystalline quality compared with layers grown on c-sapphire.\textsuperscript{[46]} ZnO therefore seems to be the more suitable choice as back contact.

For a deeper investigation on the growth of the κ-(Al\textsubscript{x}Ga\textsubscript{1−x})\textsubscript{2}O\textsubscript{3} layers on the ZnO templates, RSM measurements were carried out around the asymmetric (139) reflection of the alloy layers and the (10.5) reflection of ZnO. Figure 3a shows RSMs for selected values of x. The ZnO (10.5) reflection exhibits low broadening both in q\textsubscript{∥} and in q\textsubscript{⊥}, confirming the high crystalline quality of the back contact layer as growth template. The (139) reflection is not aligned in q\textsubscript{∥} with the ZnO (10.5) reflection such that pseudomorphically strained growth can be excluded, which would also not be expected at the given values of the lattice mismatch and the large film thicknesses. However, the (139) reflection shows a similar low broadening as the ZnO template in q\textsubscript{⊥} independent of x in agreement with previous reports,\textsuperscript{[19,24]} corroborating the same high quality of the alloy layers compared with the growth on single-crystalline c-sapphire substrates. The main broadening of the reflections is given along the q\textsubscript{∥} direction, indicating limited lateral regions of coherent scattering of the X-rays as the cause, i.e., the size of the rotational domains as limiting factor. This also confirms the same interpretation of the larger broadening in the symmetric rocking curves as discussed earlier. For mosaicity being the limiting factor, the peaks have to be broadened mainly in the radial direction given by the connection of the center of the coordinate system and the respective peak. The broadening itself is further almost independent of x, but the direction of the broadening slightly tilts exactly toward the aforementioned axis, indicating an increasing mosaicity with increasing x, as observed for the growth on c-sapphire substrates as well.\textsuperscript{[24]}

The position of the (139) reflections in reciprocal space shifts to larger values in q\textsubscript{∥} as well as q\textsubscript{⊥} with increasing x equivalent to a reduction both in the out-of-plane c-lattice constant already discussed earlier and in the in-plane d\textsubscript{130} lattice plane distance with increasing x. We determined these values from the d\textsubscript{∥} and d\textsubscript{⊥} positions of the (139) reflection in reciprocal space, which were corrected by the position of the ZnO (10.5) reflection to account for sample tilts. Figure 3b shows the determined values of d\textsubscript{130} and c as a function of x. Both decrease linearly in agreement with Vegard’s law. The evolution of the c-lattice constant follows the relation determined from the XRD 2θ–ω scans as given by Equation (1) with good agreement (purple dashed line). From a linear fit to the data for the d\textsubscript{130} in-plane lattice plane distance, the following relation is obtained

\[
d_{130}(x) (\text{Å}) = (2.511 \pm 0.002) - (0.112 \pm 0.005)x
\]

The magnitude of the slope is larger than the one obtained for the growth on c-sapphire substrates (−0.074Å in a previous study\textsuperscript{[24]}), which might be due to several reasons. First, the

![Figure 3](image-url)

Figure 3. a) Typical RSMs around the asymmetric κ-(Al\textsubscript{x}Ga\textsubscript{1−x})\textsubscript{2}O\textsubscript{3} (139) and ZnO (10.5) reflection for selected Al contents x as indicated. For the sample with x = 0.48, the additionally needed κ-Ga2O3 template layer causes the additional reflection. The weak reflection at q\textsubscript{∥} ≈ 9.9 nm\textsuperscript{−1} for x = 0 is caused by gold contacts on the layer that were not in the measured regions for the other samples and fit well to the position of the Au (331) reflection (JCPDS card no. 04-0784) in 2θ as well as ω when (111) oriented growth is considered. b) c-lattice constant and in-plane lattice plane distance d\textsubscript{130} as a function of x determined from the RSM measurements as indicated. The green dashed line is a fit to the d\textsubscript{130} data with the given relationship. The purple dashed line is no fit to the c-lattice constants, but the relation c(x) according to Equation (1) as determined from the XRD 2θ–ω scans.
reflections are broadened in \( q_x \), which causes the determination of the position of the reflection in this coordinate to be more erroneous in general; the errors given in Equation (2) are merely the ones given by the fit. Second, the correction of the position of the reflections in reciprocal space using the position of the ZnO (10.5) reflection might introduce additional systematic errors, as the thin-film lattice constants might deviate from the single-crystal data on ZnO. We could use the actual \( c \)-lattice constant of the ZnO thin film determined from the XRD 2θ-ω scans for \( q_x \), but for the in-plane \( a \)-lattice constant, we needed to refer to literature values (JCPDS card no. 79-2205). Unfortunately, no substrate reflection was available at the given position in \( q_x \), which is typically used for correction. Third, the transfer factor of Al from the target to the thin film layer for the given samples was only \( x \) (film)/\( x \) (target) \( \approx 1.4 \), which is lower than that for the growth on bare c-sapphire substrates using the same targets, which was \( \approx 1.6 \) in previous reports at the same growth conditions.\(^{[24,26]}\)

The ZnO/ZnO:Al template might change the actual growth temperature due to a different equilibrium with the radiation field of the resistive heater or changes the incorporation of Al in general. A difference in growth temperature influences both Al incorporation, which decreases with decreasing \( T_g \) due to the lower amount of volatile \( \text{Ga}_2\text{O} \) suboxides that are formed and desorbed,\(^{[64]}\) and the unit cell volume in general that could change the evolution of in-plane lattice constants. Regarding all these factors, the relation according to Equation (2) is in agreement with the one obtained in a previous study.\(^{[24]}\)

The \( \kappa-(\text{Al},\text{Ga}_{1-x})_2\text{O}_3 \) layers on the ZnO/ZnO:Al therefore show similar growth behavior and feature the same crystallographic characteristics and crystal quality as for single-crystalline substrates, such that the growth templates can be implemented as back contact layers in device applications using active \( \kappa \)-phase structures. However, for such devices, the surface morphology is of great importance, which will be investigated in the next section.

### 2.3. Surface Morphology

To evaluate the surface morphology, AFM images were recorded for all samples. Figure 4 shows the AFM images for different Al contents \( x \) in comparison with a bare ZnO/ZnO:Al template layer. The typical granular nature of the thin films with grain sizes below 100 nm supposedly caused by the three rotational domains is clearly observable. The determined roughness values are in the low nm regime and the incorporation of Al seems to result in smoother surfaces than for the binary layers. Slightly different general appearances of the surface morphologies considering grain sizes and grain size distributions are similar to those observed before by us for a \( \kappa-(\text{Al},\text{Ga}_{1-x})_2\text{O}_3 \) thin film with a lateral composition gradient on bare c-sapphire substrates\(^{[25]}\) and are not consequences of the growth on the ZnO buffer layer. This might be caused by the different Al contents or inherent variations of the PLD process in general. Other possible reasons are the different targets used for each \( x \) that were fabricated in our laboratory or the effect of the tin-enriched surface layer as discussed later. Otherwise, there is no coherent trend regarding the roughness values which stay always in the range of \( R_q \approx 2 \) nm independent of \( x \). The obtained roughness values are slightly larger than that observed before on c-sapphire substrates,\(^{[24]}\) which might be caused by the initial roughness of the template layers that is already given with \( R_q = 1.7 \) nm, as ZnO tends to grow in a 3D growth mode.\(^{[58,65]}\) Nevertheless, these roughness values are sufficient for the application of the layers in device structures. Further, as discussed in a different report,\(^{[66]}\) the surface morphology of \( \kappa \)-phase layers might not reflect the actual interface roughness within more sophisticated heterostructures such as SLs. This could be due to the proposed tin-assisted growth mechanism,\(^{[19]}\) where a liquid tin alloy layer is floating on top of the growing thin-film surface. The surface morphology therefore might be only determined by the tin rich surface layer found by us in depth-resolved X-ray photoelectron spectroscopy studies.\(^{[19]}\) The actual interface roughnesses can therefore be much lower, as proven by narrow SL fringes in XRD patterns up to high orders for SL heterostructures in a previous report by us.\(^{[26]}\)

#### Figure 4.

Typical surface morphologies as given by AFM images for the bare ZnO/ZnO:Al template layers and for the \( \kappa-(\text{Al},\text{Ga}_{1-x})_2\text{O}_3 \) thin films on the templates with different values of \( x \) as indicated. \( R_q \) denotes the root-mean-squared surface roughness of the samples determined from the images. The surface roughness is mainly determined by the roughness of the ZnO template layer.

To prove the applicability of Al-doped ZnO as back contact layer and growth template for sophisticated heterostructure devices
based on $\kappa-(\text{Al}_{0.27}\text{Ga}_{0.73})_{0.48}$O$_3$ / $\kappa-\text{Ga}_2\text{O}_3$ heterostructures, we grew a 15-layer pair QW SL heterostructure on a ZnO:Al template with $\kappa-(\text{Al}_{0.27}\text{Ga}_{0.73})_{0.48}$O$_3$ as barrier and $\kappa-\text{Ga}_2\text{O}_3$ as QW layer with nominal thicknesses of 20 and 3 nm within each layer pair, respectively. A complete description of the structure is found in the Experimental Section. Figure 5a shows the XRD 2$\theta$–$\omega$ scan of the SL structure. Apart from the substrate and template layer reflections, only the zero order reflections of the SL structure at the Bragg reflection angles corresponding to the average c-lattice constant of the structure and adjacent SL fringes are observable. The first ones are indicated with SL$_0$ (00n) and are close to the (00n) reflections of the $\kappa-(\text{Al}_{0.27}\text{Ga}_{0.73})_{0.48}$O$_3$ barrier. SL fringes are marked with a green asterisk. Sharp and intense SL fringes occur up to high orders, in this case up to the fifth with respect to the SL$_0$ (002) reflection, and are observed up to the SL$_0$ (0010) reflection. The fringes show a broadening in the order of 0.07° 2$\theta$ similar to SL structures grown on c-sapphire substrates. The occurrence of the SL fringes up to high orders and up to large angular positions as well as their high intensity and low broadening confirms a high crystalline quality and smooth interfaces with necessary roughnesses of only a few monolayers across the whole stack. Any increased interface roughnesses would increase the broadening and diminish the intensity of the SL fringes. From the separation of the SL fringes, the double-layer thickness, the average combined thickness of one barrier and one QW layer in the SL structure, can be determined to be $\approx$23.2 nm, agreeing well with the design of the SL. The growth of the SL occurs in three rotational domains as well and epitaxial relations to the ZnO:Al template are similar to those given earlier. Corresponding XRD $\phi$ scans can be found in the Supporting Information.

Sharp SL fringes up to the fifth order can also be observed around the SL$_0$ (139) reflections in the RSM shown in Figure 5b. Moreover, the SL fringes and SL$_0$ (139) are aligned in $q_\parallel$, indicating coherent growth of all layers in the SL structure.

The surface morphology of the SL structure depicted as an AFM image in Figure 5c shows a similar nanocrystalline growth as the single alloy layer in Figure 4. The roughness with $R_q = 1.3$ nm is slightly lower than that for the single layer with a comparable Al content, which is due to the missing unintentionally doped ZnO buffer layer and corroborates the interpretation earlier. Nevertheless, it is still much larger than the interface roughness of a few monolayers that can be deduced from the XRD pattern. Therefore, the surface morphology indeed does not necessarily correspond to the achievable interface roughness in $\kappa$-Ga$_2$O$_3$-based heterostructures. Moreover, these results show that SL heterostructures with a high crystalline quality can be grown on a highly conductive ZnO:Al back contact layer, which can be utilized in sophisticated device applications such as QWIPs, where the SL serves as active layer.

3. Conclusion

In summary, we have deposited high-quality $\kappa-(\text{Al}_{x}\text{Ga}_{1-x})_{0.48}$O$_3$ single layers and heterostructures up to $x = 0.48$ on highly conductive Al-doped ZnO (00.1) thin-film back contact layers on...
a-sapphire substrates by PLD. The alloy layers show a similar high crystalline quality and smooth surfaces with roughness values in the low nm regime, as comparable thin films on single-crystalline substrates investigated in previous publications. The evolution of in-plane and out-of-plane lattice constants with Al content was found to be in agreement with previous reports as well. The epitaxial growth of all layers with three rotational domains on the ZnO templates was confirmed by XRD $\phi$ scans and epitaxial relationships of $k$-(AlGa)$_{1-x}$O$_2$ to the ZnO template layer were determined. RSM measurements of asymmetric $k$-(AlGa)$_{1-x}$O$_2$ (139) reflections further confirmed similar broadening and evolution of the peaks with Al content as similar layers on single-crystalline substrates. A 15-layer-pair $k$-(AlGa)$_{1-x}$O$_2$/Ga$_2$O$_3$ SL heterostructure deposited on the ZnO:Al back contact layer showed sharp and intense SL fringes up to the fifth order in XRD 2$\theta$–$\omega$ scans as well as RSM measurements indicating monolayer smooth interfaces and coherent growth of the SL structure. Smooth surface morphology could be verified in AFM images as well. These results confirm the excellent applicability of highly conductive Al-doped ZnO as a growth template and back contact layer for sophisticated devices based on $k$-(AlGa)$_{1-x}$O$_2$ heterostructures and SLs as active layers, such as QWIP applications.

4. Experimental Section

Sample Preparation: The thin-film layers of all samples were grown by PLD on 10 x 10 mm$^2$ a-sapphire substrates. The $k$-(AlGa)$_{1-x}$O$_2$ layers with a thickness of 500–600 nm were deposited on an unintentionally doped 400nm-thick ZnO buffer layer on top of a $\approx$200nm-thick heavily Al-doped ZnO back contact layer ($\rho \approx 2 \times 10^{-5}$ $\Omega$m, $n \approx 6 \times 10^{18}$ cm$^{-3}$, and $\mu \approx 50$ cm$^2$Vs$^{-1}$) as determined by Hall effect measurements) on the a-sapphire substrate. It was shown that an unintentionally doped ZnO buffer layer was needed to equilibrate the conduction bands of $\beta$-Ga$_2$O$_3$ and ZnO to achieve ohmic behavior of the whole junction.[19] Therefore, this approach has been followed in this report as well. For the layer with $x = 0.48$, an additional $\approx$150 nm-thick binary $\kappa$-Ga$_2$O$_3$ buffer layer was needed to be introduced below the $k$-(AlGa)$_{1-x}$O$_2$ alloy layer to stabilize the $k$-phase. This layer was needed in general for our layers with $x > 0.4$ as extensively investigated in a previous study.[24] The QWSL heterostructure sample consisted of 15 layer pairs of alternating binary $\kappa$-Ga$_2$O$_3$ QW layer ($\approx$3 nm) and $k$-(Al$_{0.27}$Ga$_{0.73}$)$_2$O$_3$ barrier layer ($\approx$20 nm) on top of a $\approx$100 nm $k$-(Al$_{0.27}$Ga$_{0.73}$)$_2$O$_3$ barrier layer. Here, this structure was directly grown on a $\approx$200 nm-thick heavily Al-doped ZnO back contact layer without the additional ZnO buffer layer. All sample structures are also schematically shown in Figure S2,

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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