The electrical conductivity of cubic (In$_{1-x}$Ga$_x$)$_2$O$_3$ films ($x \leq 0.18$): Native point defects, Sn-doping, and the surface electron accumulation layer

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The alloying of the group-III transparent semiconducting sesquioxides In$_2$O$_3$ and Ga$_2$O$_3$ can lead to a modulation of the properties of the parent compounds, e.g., the shallow- and deep-donor character of the oxygen vacancy or the presence and absence of a surface electron accumulation layer, respectively. In this work, we investigate the effect of alloying on the electron transport properties of unintentionally-doped single-crystalline and textured bixbyte (In$_{1-x}$Ga$_x$)$_2$O$_3$ thin films annealed in oxygen and vacuum with Ga contents up to $x = 0.18$. Hall effect measurements demonstrate a surprising increase in electron density due to native defects with added Ga. This increase may be related to the incorporation of Ga-interstitials or oxygen vacancies induced by Ga-related unit-cell distortions. A combined investigation based on hard and soft x-ray photoelectron spectroscopy measurements demonstrates the existence of the surface electron accumulation layer for all alloy films and, hence, no depletion up to $x = 0.18$. Finally, we additionally demonstrate a single-crystalline (In$_{0.92}$Ga$_{0.08}$)$_2$O$_3$:Sn film, as a possible transparent conductive oxide with a wider band gap than that of (Sn-doped) In$_2$O$_3$.

I. INTRODUCTION

In$_2$O$_3$ and Ga$_2$O$_3$ are transparent semiconducting materials widely studied for implementation in devices.

On the one hand, In$_2$O$_3$ is typically cubic bixbyte$^1$ and has an optically forbidden direct band gap found to be 2.7–2.9 eV, with strong optical absorption occurring from valence bands nearly 1 eV below the valence band maximum (VBM).$^2$–$^4$ This property renders In$_2$O$_3$ transparent in the visible range of the electromagnetic spectrum and is—remarkably—combined with high electrical conductivity. In$_2$O$_3$ exhibits inherent $n$-type conductivity, which is commonly referred to as unintentional doping (UID) and can be significantly enhanced by doping. The unintentional doping of In$_2$O$_3$—with electron concentrations ranging from $10^{17}$ to $10^{19}$ cm$^{-3}$—is typically attributed to hydrogen impurities, such as singly-charged hydrogen interstitials, $H^+_i$, and substitutional hydrogen at an oxygen site, $H_O^*$.$^5$ The origin of the unintentional conductivity of In$_2$O$_3$ has been further associated with oxygen vacancies, $V_O^{2+}$, acting as doubly-ionized shallow donors. It is still, however, debated whether the $V_O$ are indeed shallow donors$^6$–$^8$ or deep donors that do not contribute free electrons.$^5$–$^9$ The shallow donor case is also supported by experimental results showing that annealing in oxygen, and thereby removing the oxygen vacancies, decreases the electron concentration in the bulk of In$_2$O$_3$, whereas annealing the material in vacuum does not only restore the electron concentration, but it further increases it.$^{10}$ Indium interstitials, that are predicted to act as shallow donors, may also explain these experimental results but are considered unlikely due to their high formation energy.$^8$–$^9$ In$_2$O$_3$ also possesses a few-nanometers-thick, two-dimensional surface electron accumulation layer (SEAL), the existence of which has been demonstrated by a downward band bending at the surface and the presence of quantized subbands.$^{11}$–$^{13}$ The conductivity of the SEAL is a particularly important property for applications of In$_2$O$_3$ as gas-sensing material$^{14}$, facilitates the formation of ohmic contacts but makes the formation of Schottky contacts challenging.$^{15}$ Various methods allow the modulation of the SEAL: A reduction of the SEAL carriers can be achieved by an oxygen plasma treatment of the surface at elevated temperatures$^{14,16,17}$, by adsorption of oxidizing reactive species, such as $O_2$ and $O_3$$^{18}$ or by intentional compensating acceptor doping.$^{10}$ Conversely, annealing treatments$^{16,17}$ or illumination with above-bandgap light$^{14,18}$ can enhance the SEAL. Beyond the unintentional conductivity of In$_2$O$_3$, highly Sn-doped In$_2$O$_3$—commonly known as indium-tin oxide (ITO)$^{20}$–$^{21}$—is commercially used as a transparent contact in optoelectronics, such as displays, light-emitting diodes, and solar cells.

Ga$_2$O$_3$, on the other hand, has several polymorphs$^{22}$, the most stable out of which is its $\beta$-phase with a monoclinic crystal structure. It has a band gap of approximately 4.8 eV$^{24}$ and is, thus, transparent within both the visible and well within the UV range. Contrary to In$_2$O$_3$, MBE-grown Ga$_2$O$_3$ is insulating at room temperature$^{25}$ even though hydrogen has been shown to be a shallow donor in Ga$_2$O$_3$ as well.$^{26}$ As for the nature of $V_O$, it is widely accepted that they behave as deep donors in Ga$_2$O$_3$. Moreover, Ga$_2$O$_3$ typically exhibits upward band bending$^{27,28}$ and, hence, a surface electron depletion layer, providing challenges for the formation of ohmic contacts.$^{29}$

In theory, a combination of these two compounds can lead to a modulation of their original properties, such as the position in the band gap—and consequently activation energy—of the $V_O$, the surface electron density or even the presence of the SEAL altogether. Figure 1 (a) graphically illustrates the current knowledge about the relative band-edge positions of cubic In$_2$O$_3$ and $\beta$-Ga$_2$O$_3$, along with the charge state transition levels of $V_O$ with respect to the conduction band minimum (CBM). These have been extrapolated from the formation energy diagrams of Chatratin et al.$^3$ and Deák et al.$^{30}$ depicted schematically in Fig 1 (b). An increase of the $V_O$ activation energy would be particularly interesting, as it could possibly
benefit the gas sensitivity in In$_2$O$_3$, by reducing both the parallel, parasitic contribution of the bulk conductivity to that of the gas-sensitive surface and the concentration of adsorption centers. A modulation of the SEAL transport properties could also benefit this same application.

Experimentally, a decrease of the Hall electron concentration and mobility of MOCVD-grown (In$_{1-x}$Ga$_x$)$_2$O$_3$ films grown on well-matched ZrO$_2$:Y (YSZ) substrates has been earlier demonstrated by Kong et al.\textsuperscript{31} for increasing $x$ up to 0.9. However, limited information is given on the phase purity: the films of this study surprisingly remain cubic up to $x = 0.5$, which contradicts current knowledge about the transition from cubic to monoclinic phase\textsuperscript{32} and may be explained by a lower incorporation of Ga in the cubic phase than the given total Ga content. Recent investigations by Nagata et al.\textsuperscript{33} and Swallow et al.\textsuperscript{34} on films grown using pulsed layer deposition (PLD) show the existence of a SEAL up to approximately $x = 0.4$. The latter also demonstrates the lowering of the charge neutrality level with respect to the conduction band minimum from In$_2$O$_3$ to IntGaO$_3$ to Ga$_2$O$_3$ and, hence, the reduction of the donorlike defects with Ga. However, these studies do not explore the transport properties and, particularly, the electron mobility of those films. Previously, In$_2$O$_3$ (and Ga$_2$O$_3$) films grown by PLD have been shown to exhibit comparably low electron mobilities,\textsuperscript{35} likely related to high concentrations of defects.

Another interesting prospect for (In$_{1-x}$Ga$_x$)$_2$O$_3$ alloys for applications requiring transparent conductive materials with a larger band gap than that of ITO can be explored through additional doping with Sn (IGTO). Recent, application-oriented studies towards this direction have demonstrated, for instance, polycrystalline or even amorphous IGTO thin film transistor with high field-effect mobilities.\textsuperscript{36,37} IGTO has been further proposed as an alternative transparent conducting electrode for light-emitting diodes (LEDs), with the intention to increase their external quantum efficiency in the near-ultraviolet spectral range, due to its increased absorption edge, compared to ITO.\textsuperscript{38}

Recently, we have studied well-defined, high-quality, single-crystalline (In$_{1-x}$Ga$_x$)$_2$O$_3$ layers at the low-$x$ end grown by plasma-assisted molecular beam epitaxy (PA-MBE) in terms of surface and film morphology, crystalline quality, and homogeneity\textsuperscript{39} as well as increasing band gap and optical absorption edge with $x$.\textsuperscript{40}

In this work we investigate the effect of Ga on the unintentional conductivity of the bulk and the SEAL and demonstrate intentional Sn-doping.

II. EXPERIMENTAL DETAILS

For the purposes of this study, high quality (111)-oriented single-crystalline (In$_{1-x}$Ga$_x$)$_2$O$_3$ films, including one (In$_{0.92}$Ga$_{0.08}$)$_2$O$_3$:Sn film, were synthesized by PA-MBE on quarters of 2-in. insulating YSZ (111) substrates, whereas full 2-in. insulating Al$_2$O$_3$ (0001) (c-plane Al$_2$O$_3$) substrates have been employed for the growth of textured samples. The study focuses on the low-$x$ ($x \leq 0.18$) bixbyte phase end of (In$_{1-x}$Ga$_x$)$_2$O$_3$, hence the substrate choice is based on suitability for heteroepitaxy of pure In$_2$O$_3$. After growth, all samples have been further cleaved into smaller pieces with a size of approximately $5 \times 5$ mm$^2$. The total thickness of the films ranges between 310 nm (SII) and 700 nm [SIII, including the (In$_{0.92}$Ga$_{0.08}$)$_2$O$_3$:Sn film]. Detailed information on the growth of these single-crystalline samples and the determination of the Ga content therein are reported in our recent work in Ref. 40.

The films on c-Al$_2$O$_3$—not described in our previous work—have been grown under similar conditions upon a pure In$_2$O$_3$ buffer layer at the interface. The nucleation of the buffer layer was conducted under an oxygen plasma flux of 2 standard cubic centimeters per minute (SCCM), whereas the main part of the film was grown under a flux of 0.5 SCCM. The power of the oxygen plasma source was maintained at 300 W throughout the entire growth procedure. The In-cell temperature was consistently at 850°C for all sam-
samples, corresponding to a beam equivalent pressure (BEP) of $4 \times 10^{-7}$ mbar. The Ga-containing layers were grown using varying Ga-cell temperatures from 790°C up to 840°C with respective BEPs ranging between 1.1 – 3.5 $\times 10^{-8}$ mbar. Under these conditions the films developed with a growth rate of approximately 1 Å/s. The substrate temperature was maintained at 750°C throughout both the nucleation and main film growth—as measured by a thermocouple between the substrate and the substrate heater. This is significantly higher than the substrate temperature used for growth on YSZ, as wetting and overall growth is more straightforward on Al$_2$O$_3$. The oxygen flow was discontinued after the completion of the growth from 600°C downwards and the cool-down was essentially realized in vacuum. In contrast to the growth procedure on YSZ, an faster cool-down rate of 0.5°C/s was employed, as there is no concern for film delamination in the case of growth on c- Al$_2$O$_3$.

Based on the fact that the oxygen chemical potential is higher in an O-rich environment and it decreases with increasing temperature, the free electron concentrations of the films have been intentionally varied by annealing treatments in oxygen and vacuum in order to investigate the effect of Ga on transport. Decrease in the electron concentration has been achieved through exposure of the films in an oxidizing environment at high temperature, hereafter referred to as O-annealing. This treatment was performed within a rapid thermal annealing (RTA) system at a final temperature of 800°C. This treatment was performed within a rapid thermal annealing (RTA) system at a final temperature of 800°C. After annealing, the surface band bending behavior, the take-off angle (TOA: $\theta$) dependent PES was performed. TOA is an angle between the normal vector to the sample surface and the detector direction. A high angle PES is surface sensitive, while a low angle it is bulk sensitive. The corresponding inelastic mean free path (IMFP) of HAXPES and SXPES for the In 3d core-level photoemission calculated by the Tanuma–Powell–Penn-2M are $\lambda_{\text{HAXPES}} = 7.29 \text{nm}$ and $\lambda_{\text{SXPES}} = 2.39 \text{nm}$, respectively, meaning that HAXPES probes three times deeper than SXPES. The probing depth is three times the IMFP, therefore, our HAXPES measurements probe approximately 22nm below the sample surface, which can reduce the effect of the surface Fermi level pining of In$_2$O$_3$. To estimate the band bending behaviors of In$_2$O$_3$, a detailed XPS spectral analysis was performed using COMPRO (Common Data Processing System) version 11 written by Yoshihara and Yoshikawa, which simulates the potential energy distribution using the convolution of calculated peaks at several TOAs. The simulated spectra were obtained to reproduce the experimental SXPES and HAXPES spectra at several TOAs by adjusting the band bending profiles and minimizing the chi squares of the difference between the simulated and experimental spectra. The band bending direction was denoted as downward and upward, i.e., towards lower and higher binding energies, respectively.

III. RESULTS AND DISCUSSION

Figure 2 shows the transport properties of the single-crystalline and textured films, as determined by four-point Hall effect measurements in the van der Pauw arrangement.

A. Unintentional bulk doping

Electron concentration

The electron concentrations, shown in Fig. 2 (a), on the order of $\sim 10^{17}$ cm$^{-3}$ and $\sim 10^{18}$ cm$^{-3}$ for the pure In$_2$O$_3$ films (Ga content $x = 0$) annealed in oxygen and vacuum, respectively, are in good agreement with our previous findings and confirm, that an oxygen-deficiency provides unintentional shallow donors, e.g., oxygen vacancies. With increasing Ga-content, we observe a slight increase of the electron concentration of the films annealed in oxygen, which may, in principle, be related to impurities in the Ga-source that act as shallow donors in the films.
For the single-crystalline films annealed in vacuum, however, the electron concentration is significantly higher and its increase with Ga-content is significantly stronger, which cannot be explained by such impurities and, instead, should be related to point defects due to an oxygen-poor stoichiometry of the films caused by the annealing. A plausible explanation for this unanticipated increase in the Hall electron density $n_{3D(H)}$ could be the formation of donor-like native lattice defects driven by Ga-induced unit cell distortions.

As discussed, it has been shown that the native lattice defects with the lowest formation energy in In$_2$O$_3$ that can act as donors are V$_{O}$. An O-annealing treatment can potentially fill in V$_{O}$ in the films and reduce $n_{3D(H)}$. Fig. 2 (a) shows exactly this effect, if one compares the O-annealed to the vacuum-annealed state of the films. Since the degree of V$_{O}$-filling upon O-annealing is not controllable, it is not necessary that the O-annealed Ga-containing samples display a decreased $n_{3D(H)}$ compared to the undoped case. However, assuming the (+/0) transition level for V$_{O}$ suggested by Chatratin et al.\textsuperscript{8} to correspond to the maximum achievable Fermi level in In$_2$O$_3$ due to V$_{O}$ and considering the approximation of the generalized Einstein relation for degenerate and non-degenerate semiconductors of Nilsson\textsuperscript{52}, the resulting electron concentration should never exceed $6.5 \times 10^{19}$ cm$^{-3}$ [indicated by the horizontal dashed line in Fig. 2 (a)], for an effective electron mass of 0.18$m_e$ as in Ref. 53. All vacuum-annealed Ga-containing samples demonstrate equal or higher $n_{3D(H)}$ than this upper limit. Moreover, the strongly increasing $n_{3D(H)}$ with increasing Ga-content contradicts the expected lowering of the maximum free electron concentration that can be provided by V$_{O}$ due to their expected decreasing charge transition level with increasing Ga-content.

Unless this charge transition level is significantly higher than theoretically predicted by Chatratin et al.\textsuperscript{8}, the hypothesis that the $n_{3D(H)}$ increase is due to V$_{O}$ alone does not suffice and other native donor impurities need to be considered. Those with the next lowest formation energies are metal interstitials acting as triple-donors (In$_{3}^{3+}$, Ga$_{3}^{3+}$\textsuperscript{26,30,54,55}). The neutral charge state transition levels for those seem to be well within the conduction band and they, thus, serve as a conceivable explanation for the particularly high electron concentrations observed here. In addition, the smaller ionic size of Ga$^{3+}$ than In$^{3+}$ makes its incorporation as interstitial more likely, which would also be consistent with the increasing electron concentration with increasing Ga-content.

**Electron mobility**

At a first glance, the trend of decreasing mobility with increasing Ga-content visible in Fig. 2 (b) is not particularly surprising, as mobility is generally expected to decrease with increasing carrier concentration and, thus, ionized impurity concentration. However, in the case of single-crystalline Sn-doped In$_2$O$_3$ films, mobility has been reported to be higher than in the single-crystalline films studied here for the same range of electron concentrations ($n_{3D(H)}$ : $\approx 10^{18}$ – $\approx 10^{19}$ cm$^{-3}$).\textsuperscript{56} This discrepancy would be consistent with the stronger ionized impurity scattering of doubly-ionized V$_{O}^+$ in the (In,Ga)$_2$O$_3$ films compared to the singly ionized donors in the Sn-doped In$_2$O$_3$ films, theoretically modeled by Preissler et al.\textsuperscript{56}. This effect is visualized in Fig. 3, with the IGTO data point lying between the Z = 1 (Sn$^+$) and Z = 2 (V$_{O}^{2+}$). In fact, the good agreement between the experimental data from the vacuum-annealed films and the model curve corresponding doubly-ionized donors implies that the origin of the unexpectedly high electron concentration in the alloy films cannot be attributed—for the most part—to triply-ionized metal interstitials, as suggested previously. This finding indicates that the V$_{O}$ transition level is indeed significantly higher than theoretically predicted by Chatratin et al.\textsuperscript{8}. In addition, distortion in...
the homogeneity of the crystal potential is inevitable in alloys and can limit carrier transport by alloy fluctuation scattering.

Substrate choice does not seem to significantly affect the results in terms of electron concentration. However, decreased mobility is exhibited by the films grown on c-Al₂O₃, which happens due to scattering at the grain boundaries of the rotational domains and is not an effect of the Ga incorporation.

B. Intentional bulk doping with Sn

An (In₀.₉₂Ga₀.₀₈)₂O₃:Sn film marked as IGTO in Fig. 2 has been realized as a potential advancement of ITO, with the intention of achieving both comparably high conductivities due to high electron concentrations and an increased band-gap necessary for higher photon energy applications. The enlargement of the band-gap of (In₁₋ₓGaₓ)₂O₃ with and without the addition of Sn in comparison to pure In₂O₃ has been confirmed and is discussed in detail in Ref. 41. In this work we investigate its transport properties, which can be directly compared to the (In₀.₉₂Ga₀.₀₈)₂O₃ sample with the same Ga content. As shown in Fig. 2 (a), incorporation of Sn dramatically increases the n_{3D} of the film by more than two orders of magnitude. Doping In₂O₃ and—hence—cubic (In₁₋ₓGaₓ)₂O₃ at the low-x end—with an Sn⁴⁺ ion should provide one free electron for conduction. The doping effect of Sn in ITO, however, can be largely compensated upon O-annealing treatments, as the ones performed on the films in this study, due to the formation of Sn—oxygen-interstitial clusters, 2SnO₂. Interstitial oxygen atoms can be removed under reducing conditions, which breaks down those complexes and frees two donors per O₂ in the process. Consistent with this defect model, we observed an increase of the electron concentration from 1.5 × 10²⁰ to 3.3 × 10²⁰ cm⁻³ when comparing the Sn-doped film annealed in oxygen to that annealed in vacuum. Together with the simultaneous increase of electron mobility from 38 to 51 cm²/Vs [shown in Fig. 2 (b)], the film resistivity decreases from 1.1 × 10⁻³ to 3.7 × 10⁻⁴ Ω cm, well into the range typically observed for the regime of ITO, rendering IGTO films a suitable candidate for TCO applications.

C. SEAL presence in the (In₁₋ₓGaₓ)₂O₃ films

Magnified SX-PES valence band spectra of three representative samples with Ga content of x = 0.00, 0.11, and 0.18 (Fig. 4 (a)) reveal electronic states at the Fermi level (binding energy of 0 eV), indicating the presence of the SEAL. Moreover, In 3d core level spectra obtained by SX-PES demonstrate a shift towards higher binding energies than those obtained by XPS, as compared in Fig. 4 (b). These results suggest a downward band bending at the surface, a necessary precondition for the formation of the SEAL. To identify the degree of the band bending, a detailed spectral analysis using COMPRO has been performed on the In 3d spectra obtained by SX-PES and HAXPES at various TOA. Fig. 4 (c) shows potential curve at the surface of the pure In₂O₃ film. A drastic potential change has been confirmed within 6 nm from the surface. All alloy films show similar conduction band states around the Fermi level and core level shift, which clearly demonstrates the existence of a SEAL even for samples with Ga cation contents as high as x = 0.18.

IV. CONCLUSION

Based on current knowledge on the (+/0) charge transition level of oxygen vacancies in In₂O₃ and Ga₂O₃ and their consequent behavior as shallow and deep donors, respectively, we have anticipated a decrease in unintentional donor concentration upon alloying In₂O₃ with Ga. Nevertheless, the unintentional electron concentration of the (In₁₋ₓGaₓ)₂O₃ alloy films is experimentally shown to increase with Ga—particularly for films annealed in vacuum, up to values of 2 × 10¹⁹ cm⁻³. Hence, we attribute the increased electron concentration to shallow-donorlike native lattice defects, such as oxygen vacancies or metal—especially Ga—interstitials. According to the reported (+/0) charge transition level of the oxygen vacancy in the band gap of In₂O₃, this defect cannot be the sole factor contributing to this effect, as in that case we would not observe electron concentrations higher than 6.5 × 10¹⁸ cm⁻³. Therefore, either the enhanced carrier concentration is at least partly due to Ga interstitials or unit cell distortion due to Ga-incorporation induces an increased incorporation of oxygen vacancies, whose (+/0) charge transition level is higher than reported in Ref. 8. Moreover, the mobility of the alloy films indicates their conductivity originates from doubly-ionized donors, such as oxygen vacancies, rather than triply-ionized ones, as the metal interstitials. Hence, the theoretically calculated charge transition level of the oxygen vacancies needs to be reconsidered. In addition, the surface elec-
tron accumulation layer native to In$_2$O$_3$, but not present in Ga$_2$O$_3$, does not seem to be affected by the addition Ga, according to surface-sensitive x-ray photoelectron spectroscopy measurements. Finally, intentional doping of the alloy films with Sn has been demonstrated to result in high electron conductivity and mobilities around $5 \times 10^{20}$ cm$^{-3}$ and mobilities around $50$ cm$^2$/Vs. This corresponds to an electrical resistivity of $\rho = 3.5 \times 10^{-4}$ $\Omega$cm, which is comparable to that of pure ITO.

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