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Polycrystalline κ -Ga₂O₃ on Si(100) substrates with GZO buffer layers

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Kappa-phase gallium oxide (κ -Ga₂O₃) is an emerging piezoelectric semiconductor with potential applications in radio-frequency devices. However, heteroepitaxial growth of κ -Ga₂O₃ on silicon substrates remains challenging owing to large lattice mismatch and interfacial oxidation. This study demonstrates the growth of polycrystalline κ -Ga₂O₃ thin films deposited by mist chemical vapor deposition (mist CVD) on Si(100) substrates using Ga-doped ZnO (GZO) buffer layers. Structural characterization via X-ray diffraction, scanning electron microscopy, and transmission electron microscopy reveals that κ -Ga₂O₃ films exhibit a *c*-axis oriented polycrystalline structure with random in-plane orientations, which yields isotropic properties that are advantageous for device applications. A ZnGa₂O₄ intermediate layer is identified at the κ -Ga₂O₃/GZO interface, which plays a critical role in phase stabilization. These findings indicate that polycrystalline κ -Ga₂O₃ on Si substrates represents a promising platform for piezoelectric semiconductor devices.

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Introduction

Ultra-wide bandgap (UWBG) semiconductors with bandgaps exceeding 4 eV have gained traction for use in high-power, high-frequency, and high-temperature electronic devices. Among these materials, gallium oxide (Ga₂O₃) has emerged as a promising candidate owing to its excellent material properties, including a wide bandgap of approximately 4.9 eV and high breakdown electric field exceeding 8 MV cm⁻¹.¹ Ga₂O₃ exists in five distinct polymorphs: α , β , γ , δ , and κ (also referred to as ε). The β -phase is the most thermodynamically stable and has been extensively examined for power device applications.^{2,3} By contrast, the metastable phases, particularly $\kappa(\varepsilon)$ -Ga₂O₃, have received relatively limited attention despite their distinctive crystal structures and functional properties.

The $\kappa(\varepsilon)$ -Ga₂O₃ polymorph crystallizes in an orthorhombic structure (space group *Pna2*₁) and is predicted to exhibit strong spontaneous polarization along the *c*-axis,⁴ making it a promising material for piezoelectric applications such as high-electron-mobility transistors (HEMTs) and surface acoustic wave (SAW) devices.^{5,6} Chen *et al.* recently demonstrated strong

piezoelectricity in ε -Ga₂O₃ thin films with $d_{33} \approx 10.8$ – 11.2 pm V⁻¹—approximately twice that of AlN—and successfully fabricated SAW resonators operating in the GHz range, making κ -Ga₂O₃ a promising candidate for radio-frequency applications.⁷ The pseudo-hexagonal arrangement of oxygen atoms on the κ -Ga₂O₃(001) surface enables heteroepitaxial growth on substrates with hexagonal or pseudo-hexagonal symmetry. High-quality epitaxial κ -Ga₂O₃ films have been fabricated on diverse single-crystal substrates^{4,8–12} employing diverse deposition techniques including halide vapor phase epitaxy (HVPE),⁸ metal-organic chemical vapor deposition (MOCVD),⁹ mist chemical vapor deposition (mist CVD),^{10–12} molecular beam epitaxy (MBE),¹³ and pulsed laser deposition (PLD).¹⁴ These epitaxial films typically exhibit characteristic 120° rotational domain structures resulting from the pseudo-hexagonal symmetry of κ -Ga₂O₃(001) surface, as observed by Cora *et al.* via plan-view TEM analysis.¹⁵

Although these single-crystal substrates have supported foundational studies and proof-of-concept devices, integration with silicon substrates is essential for practical applications owing to cost, scalability, and CMOS compatibility. Prior studies demonstrated κ -Ga₂O₃ growth on Si using epitaxial buffer layers, such as AlN or Mo,^{16,17} which can form single-crystal templates on Si despite the large lattice mismatch.

For practical piezoelectric device applications, crystallographic isotropy is crucial for design flexibility, as anisotropic properties can limit device orientation and layout options. These ordered 120° rotational domains can provide partial isotropy by averaging anisotropic elastic properties over three discrete orientations.¹⁸ Building on this observation, an

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alternative polycrystalline approach using fully randomized films with [001] texture but random in-plane orientations can yield more complete isotropy by averaging over all in-plane orientations. One approach to forming polycrystalline Ga₂O₃ is to deposit a thin film at low temperature followed by post-deposition thermal annealing.¹⁹ However, because κ -Ga₂O₃ is a metastable phase, achieving single-phase growth remains challenging. To address this issue, polycrystalline κ -Ga₂O₃ films can instead be fabricated using polycrystalline buffer layers that naturally develop *c*-axis texture on amorphous SiO₂/Si interfaces, thereby promoting phase stabilization during growth. However, key questions remain regarding phase stability, structural uniformity, and interfacial reactions in polycrystalline κ -Ga₂O₃ on Si substrates. To address these open questions, this study presents, for the first time, polycrystalline κ -Ga₂O₃ growth on Si(100) using a GZO buffer layer—an approach that offers a simpler alternative to epitaxial methods employing AlN or Mo buffer layers and inherently yields isotropic film properties advantageous for piezoelectric device applications.

Irrespective of the growth approach, interface control is critical for achieving phase-pure κ -Ga₂O₃. During high-temperature deposition, unintended interfacial reactions can generate β -Ga₂O₃ or other secondary phases, which can degrade piezoelectric properties.²⁰ Therefore, the choice of buffer layer material and its chemical compatibility with both the substrate and κ -Ga₂O₃ overlayer are crucial. Zinc oxide (ZnO) and related materials, with their hexagonal wurtzite structure, represent promising candidates for templating *c*-axis-oriented κ -Ga₂O₃ growth.

In this study, we investigate the growth of κ -Ga₂O₃ thin films deposited by mist chemical vapor deposition (mist CVD) on Si(100) substrates using Ga-doped ZnO (GZO) buffer layers. The mist CVD technique offers several advantages, including low-cost equipment, atmospheric pressure operation, and high sensitivity to substrate surface conditions, which enables selective nucleation and growth. Furthermore, mist CVD demonstrates a strong compatibility with the formation and stabilization of metastable Ga₂O₃ phases.^{21,22} The choice of GZO as a buffer layer is motivated by its hexagonal wurtzite structure and the potential for *c*-axis-oriented growth, which can provide a suitable template for κ -Ga₂O₃ deposition. In addition, due to the presence of the amorphous SiO₂ layer at the Si interface, GZO is expected to grow as an in-plane randomly oriented polycrystalline film. Consequently, κ -Ga₂O₃ formed on the GZO/Si template may inherit this in-plane structural disorder, resulting in polycrystalline growth. This is expected to result in effectively isotropic behaviour in the SAW device, as polycrystalline thin films generally exhibit a higher degree of in-plane isotropy than conventionally grown epitaxial films due to the random orientation of grains.

We systematically characterize the structural properties of the resulting κ -Ga₂O₃ films *via* X-ray diffraction (XRD), and transmission electron microscopy (TEM) is employed to elucidate the crystal structure, phase composition, and interfacial characteristics. Additionally, scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectroscopy

(EDX) were performed to analyse the elemental composition of the sample. The insights gained from this study contribute to a better understanding of κ -Ga₂O₃ growth mechanisms on Si substrates and the potential of polycrystalline κ -Ga₂O₃ for piezoelectric device applications.

Experimental section

First, a Ga-doped ZnO (GZO) buffer layer was deposited onto a Si (100) substrate with a native oxide layer using RF magnetron sputtering. A 4-inch GZO target with a composition of ZnO : Ga₂O₃ = 94.3 : 5.7 wt% was applied for the deposition. Prior to sputtering, the substrate was preheated at 150 °C for 1 hour to remove residual moisture and enhance film adhesion. During the deposition, high-purity argon gas was introduced into the chamber at a flow rate of 30 sccm as the working gas, regulated by a mass flow controller. The sputtering pressure, RF power, and substrate temperature were maintained at 1 Pa, 60 W, and 150 °C, respectively. The resulting GZO film thickness was approximately 350 nm after 102 min of sputtering.

Next, κ -Ga₂O₃ thin films were grown on GZO/Si (100) substrates by mist CVD. The precursor solution comprised of gallium acetylacetonate (Ga(C₅H₇O)₃) dissolved in deionized water with 1% hydrochloric acid (HCl), where HCl was introduced to ensure complete dissolution of the compound. The precursor solution was atomized using ultrasonic transducers operating at 2.4 MHz and transported to the substrate using N₂ carrier gas. The film deposition temperature was kept at 760 °C, and deposition time was 30 min. The Ga concentration in the precursor solution was 0.025 mol L⁻¹, and the N₂ flow rate was set to 7.4 L min⁻¹. These conditions enable precise control of the growth rate and contribute to the improvement of crystalline quality.²³

X-ray diffraction (XRD, Bruker D8 Discover) measurements were performed using a CuK α (λ = 1.5405 Å) operating at 40 kV and 40 mA as the X-ray source to investigate the crystal structure and orientation relationship of κ -Ga₂O₃. Furthermore, cross-sectional and plan-view transmission electron microscopy (TEM, FEI TecnaiF20X) observations, including selected area electron diffraction (SAED) patterns and high-resolution imaging, were performed to examine microstructure and interfacial characteristics, and the measurements were conducted by Ion Technology Center Co., Ltd.

The SAED pattern was obtained using a selected-area aperture with a diameter of 200 nm. Additionally, scanning transmission electron microscopy (STEM, JEOL JEM-2100F) and energy-dispersive X-ray spectroscopy (EDX, JEOL EX-24063JCT) were performed to analyse the elemental composition of the sample.

Results and discussion

We first investigated the crystal structure and orientation relationships of κ -Ga₂O₃ thin film grown on GZO/Si(100) substrate *via* X-ray diffraction measurements. Fig. 1(a) shows the XRD 2θ - ω scan of the as-grown sample. The diffraction peaks at 2θ = 38.9° and 59.9° correspond to (004) and (006) reflections of κ -



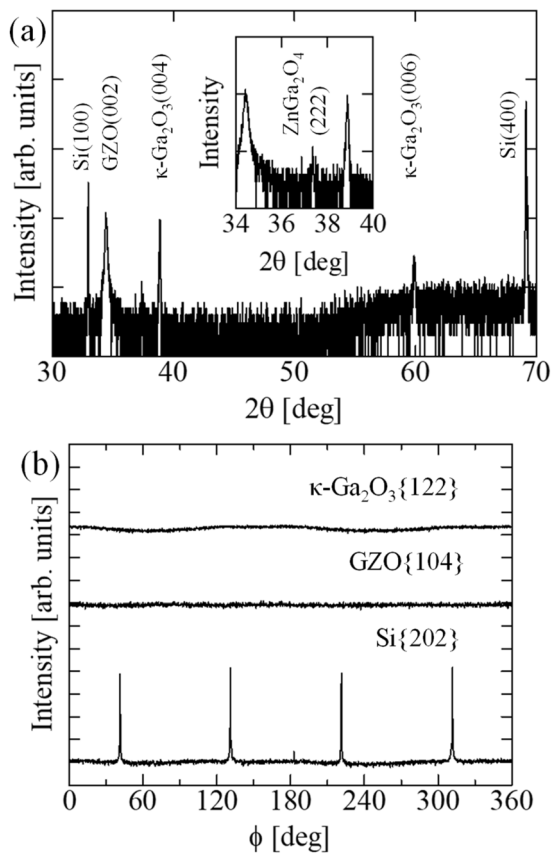


Fig. 1 (a) XRD 2θ - ω scan of κ -Ga₂O₃ grown on GZO/Si. (b) XRD ϕ -scan of three layers.

Ga₂O₃, respectively, indicating that [001]-oriented growth on the GZO/Si(100) substrate. The diffraction peak at $2\theta = 34.4^\circ$ and $2\theta = 33.0^\circ$ correspond to GZO (002) and Si(100) reflections, respectively. The presence of only (00l) reflections for κ -Ga₂O₃ and GZO confirms that both layers have preferential *c*-axis orientation perpendicular to the substrate. Notably, a small peak at $2\theta = 37.4^\circ$ is observed, corresponding to ZnGa₂O₄ (222). This suggests the formation of a ZnGa₂O₄ intermediate layer between GZO and κ -Ga₂O₃, which will be discussed in detail below based on TEM observations. Furthermore, the absence of diffraction peaks corresponding to other Ga₂O₃ polymorphs indicates that κ -Ga₂O₃ was grown in a single-phase form.

To investigate the in-plane orientation relationships, ϕ -scans measurements were performed for κ -Ga₂O₃{122}, GZO {104}, and Si{202} reflections, as shown in Fig. 1(b). The Si{202} reflection exhibits four peaks separated by 90° . By contrast, ϕ -scans of GZO{104} and κ -Ga₂O₃{122} show no characteristic peaks. With respect to GZO, this absence of peaks indicates that the buffer layer comprises *c*-axis-oriented polycrystalline grains with random in-plane orientation, which is expected for ZnO-type films grown on amorphous SiO₂ native oxide on Si substrate. Similarly, the κ -Ga₂O₃ {122} ϕ -scan shows no characteristic peaks, despite the fact that epitaxial κ -Ga₂O₃ films typically exhibit either 4 or 12 distinct peaks depending on the presence of rotation domains.^{14,24} This indicates that the κ -Ga₂O₃ thin film also comprises randomly oriented grains in the

film plane. The polycrystalline nature of GZO and κ -Ga₂O₃ layers suggests that the in-plane epitaxial relationship is not established owing to the presence of amorphous SiO₂ at the Si interface. Although the GZO buffer layer provides a hexagonally arranged oxygen template for *c*-axis-oriented nucleation of κ -Ga₂O₃, it does not define a specific in-plane orientation. These results demonstrate that κ -Ga₂O₃ grown on polycrystalline GZO(001) buffer layers exhibits a highly textured polycrystalline structure, with a preferential [001] orientation perpendicular to the substrate but random in-plane orientations.

To further investigate the polycrystalline structure and the layer formation mechanisms, we performed detailed TEM observations. Fig. 2(a) shows a cross-sectional TEM image of κ -Ga₂O₃/GZO/Si(100) structure. The thicknesses of κ -Ga₂O₃ and GZO were 320 and 360 nm, respectively. A transition layer with distinct bright contrast is observed between the GZO and κ -Ga₂O₃ layers, with a thickness of approximately 110 nm. As the κ -Ga₂O₃ layer grows away from this transition layer, a characteristic columnar structure develops, which is associated with the growth of polycrystalline grains with [001] orientation.

Fig. 2(b) shows a high-resolution TEM image of the GZO/Si interface. A thin amorphous layer approximately 5–6-nm thick is visible at the interface, corresponding to native SiO₂ formed on the Si substrate. The GZO layer exhibits polycrystalline microstructure immediately above the amorphous interface, indicating random nucleation without a defined in-plane orientation. Although individual grains maintain *c*-axis orientation perpendicular to the substrate, their in-plane orientations are not correlated owing to the absence of crystallographic templating from the underlying amorphous SiO₂. This observation confirms that the polycrystalline nature of the GZO buffer layer originates from nucleation on the amorphous SiO₂, which prevents establishment of an epitaxial relationship with the Si(100) substrate. This is consistent with the absence of characteristic peaks in the GZO{104} ϕ -scan measurement (Fig. 1(b)).

Fig. 2(c) shows a high-resolution image of the transition layer between GZO and κ -Ga₂O₃. The lattice spacing measured in this

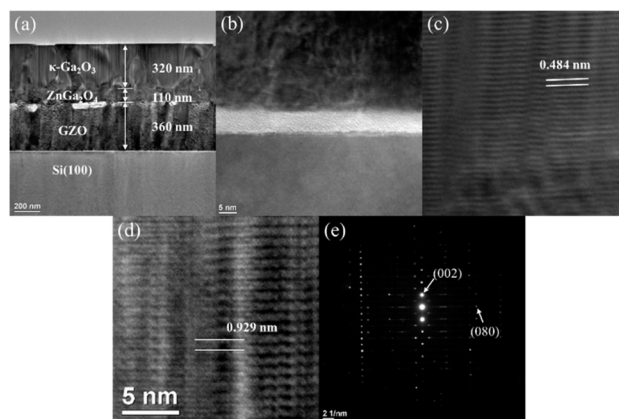


Fig. 2 (a) Cross-sectional TEM image of κ -Ga₂O₃/GZO/Si. (b) High-resolution TEM images of the GZO/Si interface, (c) ZnGa₂O₄ inter layer, and (d) κ -Ga₂O₃ film. (e) SAED pattern obtained from κ -Ga₂O₃ layer in cross-sectional view.



region is 0.484 nm, which align closely with the (111) plane spacing of ZnGa_2O_4 ($d_{111} = 0.481$ nm).²⁵ The XRD 2θ - ω peak of $\text{ZnGa}_2\text{O}_4(222)$ indicates that ZnGa_2O_4 is transiently formed at GZO interface during the growth process of κ - Ga_2O_3 . This identification is consistent with the small XRD peak observed at $2\theta = 37.4$ corresponding to $\text{ZnGa}_2\text{O}_4(222)$. The formation of intermediate layers at the interface between κ - Ga_2O_3 and underlying substrates or buffer layers is a commonly observed phenomenon. Previous studies have reported the formation of various phases, including β - Ga_2O_3 .^{15,20} The growth process of the ZnGa_2O_4 transition layer will be discussed later based on the results of STEM/EDX analysis.

Fig. 2(d) shows a high-resolution image of the κ - Ga_2O_3 layer. The lattice spacing measured adjacent lattice fringes is 0.929 nm, which aligns with the (001) plane spacing of κ - Ga_2O_3 ($d_{001} = 0.928$ nm).²⁶

Fig. 2(e) shows the SAED pattern obtained from κ - Ga_2O_3 layer in a cross-sectional view. The diffraction spots are aligned along the [001] direction, forming a single-crystal-like pattern. This appearance is attributed to the limited sample volume probed by the selected area aperture in the cross-sectional geometry, which contains only a small number of columnar grains with similar [001] orientations perpendicular to the substrate. The cross-sectional SAED primarily reflects the out-of-plane texture rather than the in-plane orientation distribution. A more definitive analysis of the in-plane structure, requiring sampling a larger area multiple grains, is provided by plan-view TEM observations discussed below.

Next, the elemental composition of the sample was investigated using STEM/EDX. Fig. 3(a)–(e) shows a scanning transmission electron microscopy (STEM) image of the κ - Ga_2O_3 /GZO/Si structure and the corresponding elemental maps of Si, Zn, O, and Ga obtained by EDX. As shown in Fig. 3(c), a region with a clearly lower Zn concentration compared with the GZO layer is observed, which corresponds to the ZnGa_2O_4 transition layer.

The formation of the ZnGa_2O_4 intermediate layer is likely attributed to the reaction between the GZO surface and HCl-containing precursor solution during the mist CVD process. The HCl likely etches the GZO surface, creating a Zn–Ga mixed

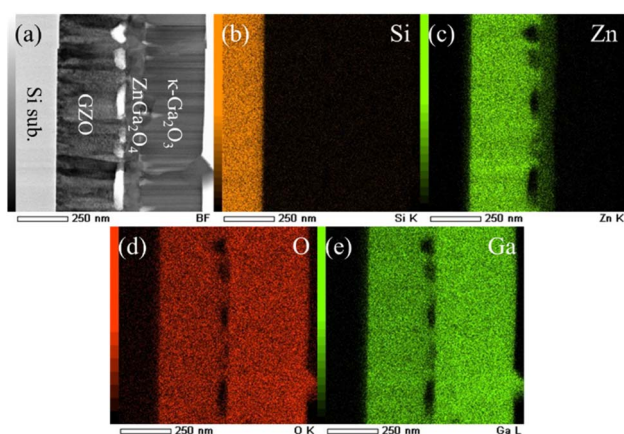


Fig. 3 (a) STEM image of κ - Ga_2O_3 /GZO/Si. (b)–(e) Elemental mapping showing the dispersion of Si, Zn, O, Ga.

composition that crystallizes as the spinel-structured ZnGa_2O_4 with (111) planes parallel to the substrate. Fig. 4 shows the oxygen atomic arrangements of κ - $\text{Ga}_2\text{O}_3(001)$ and $\text{ZnO}(0001)$, together with the atomic configuration of $\text{ZnGa}_2\text{O}_4(111)$. The $\text{ZnGa}_2\text{O}_4(111)$ plane, with its hexagonal atomic arrangement, is likely to maintain compatibility with the hexagonally arranged oxygen atoms on the underlying GZO(001) and overlying κ - $\text{Ga}_2\text{O}_3(001)$, potentially allowing the layer sequence to form without significant structural disruption. The complete orientation relationship is κ - $\text{Ga}_2\text{O}_3(001)//\text{ZnGa}_2\text{O}_4(111)//\text{GZO}(001)//\text{Si}(100)$, where the double slashes indicate parallel planes but not necessarily epitaxial relationships owing to the polycrystalline nature of the layers.

To directly investigate the in-plane orientation distribution, plan-view TEM observations were performed. Fig. 5(a) shows a plan-view TEM image, revealing a polycrystalline microstructure with multiple grains exhibiting different contrasts. The variations in contrast across the image indicate that different grains have different crystallographic orientations, with each grain diffracting differently under the same imaging conditions.

Fig. 5(b) shows a high-resolution plan-view image, where clear lattice fringes are visible from multiple grains. Importantly, adjacent grains display lattice fringes with distinctly different orientations, and the angular relationships between grains do not follow the characteristic 120 rotational pattern observed in epitaxial κ - Ga_2O_3 films. This direct observation at the atomic scale confirms that the film consists of randomly oriented polycrystalline grains rather than an ordered rotational domain structure.

Fig. 5(c) shows the SAED pattern obtained from the plan-view TEM images (Fig. 5(a)). In contrast to the single-crystal-like pattern observed in cross-sectional SAED (Fig. 2(e)), the plan-view SAED pattern exhibits circular diffraction rings rather than discrete spots. This circular pattern is the definitive signature of polycrystalline materials with randomly oriented grains and provides direct evidence that the κ - Ga_2O_3 films comprises grains with random in-plane orientations, despite the strong [001] out-of-plane texture confirmed by XRD and cross-sectional TEM. The combination of high-resolution imaging and SAED unambiguously demonstrates the polycrystalline nature of the films.

This polycrystalline structure with random in-plane orientations contrasts sharply with epitaxial κ - Ga_2O_3 films grown on

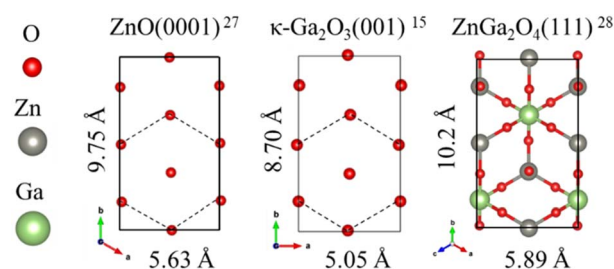


Fig. 4 The oxygen atomic arrangements of κ - $\text{Ga}_2\text{O}_3(001)$ ¹⁵ and $\text{ZnO}(0001)$,²⁷ together with the atomic configuration of $\text{ZnGa}_2\text{O}_4(111)$.²⁸



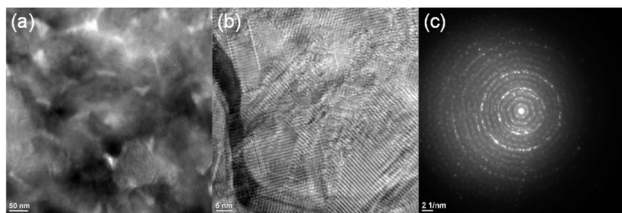


Fig. 5 (a) Plan-view TEM image of κ -Ga₂O₃ film. (b) High-resolution plan-view TEM image of κ -Ga₂O₃. (c) SAED pattern obtained from the plan-view TEM image.

single-crystal substrates, such as sapphire. Epitaxial κ -Ga₂O₃ typically exhibit well-defined in-plane orientations with characteristics 120 rotational domains resulting from the pseudo-hexagonal oxygen arrangement of κ -Ga₂O₃(001). The epitaxial films shows 12 discrete peaks in XRD ϕ -scan {122}, corresponding to three sets of four-fold symmetric domains rotated by 120° relative to each other. In exceptional cases, single-domain epitaxial growth with only four ϕ -scan peaks from {122} has been achieved on ϵ -GaFeO₃ substrates, exhibiting the same crystal structure of κ -Ga₂O₃ with small lattice mismatches. By contrast, the κ -Ga₂O₃ film in this study exhibits neither discrete ϕ -scan peaks nor discrete SAED spots or ordered lattice orientations in plan-view TEM; however, instead it shows a continuous distribution of in-plane orientations characteristics of polycrystalline materials.

This highly-textured polycrystalline structure originates from the growth on polycrystalline GZO (001) buffer layers. As observed in Fig. 2(b), the GZO layer nucleates with random in-plane orientations on the amorphous SiO₂ at the Si interface. Each GZO grain maintains *c*-axis orientation perpendicular to the substrate but has a different azimuthal orientation. The κ -Ga₂O₃ film likely grows epitaxially on individual GZO grains, with each κ -Ga₂O₃ grain through the ZnGa₂O₄ intermediate layer, as evidenced by the columnar structure observed in cross-sectional TEM. However, given that the GZO grains have random azimuthal orientations, the κ -Ga₂O₃ grains that form on different GZO grains also exhibit random in-plane orientations, resulting in the observed polycrystalline structure with strong [001] out-of-plane texture but random in-plane orientations. This structure differs fundamentally from epitaxial growth on single-crystal substrates, where the substrate defines a unique in-plane orientation for all grains, enabling the formation of ordered rotational domain structures across the entire film.

Since ZnGa₂O₄ does not exhibit piezoelectric properties, its formation is expected to degrade the overall piezoelectric response of the film. However, this effect may be mitigated by reducing the thickness of the GZO buffer layer, thereby suppressing Zn diffusion into the Ga₂O₃ layer.

Conclusions

In this study, we successfully demonstrated the growth of polycrystalline κ -Ga₂O₃ thin films deposited by mist CVD on Si(100) substrates using GZO buffer layers. The *c*-axis-oriented

GZO buffer layer promotes the formation of κ -Ga₂O₃ due to its hexagonal oxygen atomic arrangement. Furthermore, the presence of amorphous SiO₂ on the Si substrate prevents the establishment of an epitaxial relationship in the κ -Ga₂O₃/GZO/Si structure, resulting in the polycrystalline growth of κ -Ga₂O₃. Unlike previous studies employing epitaxial buffer layers such as AlN or Mo, the use of a polycrystalline GZO buffer layer provides a simpler growth process and inherently isotropic film properties, making it a more practical approach for integrating κ -Ga₂O₃-based piezoelectric devices with Si technology. Structural analysis revealed that the κ -Ga₂O₃ films exhibit random crystallographic orientations, which inherently provide isotropic properties that are advantageous for piezoelectric device applications. The formation of a ZnGa₂O₄ intermediate layer at the κ -Ga₂O₃/GZO interface was identified as a key factor in stabilizing the κ -phase. Unlike conventional epitaxial κ -Ga₂O₃ films with ordered rotational domains, the polycrystalline nature of our films offers more complete isotropy and potentially simplifies the growth process on technologically important Si substrates. These results open new possibilities for integrating κ -Ga₂O₃-based piezoelectric devices with silicon technology. Further optimization of the growth conditions and buffer layer engineering can potentially enable high-performance piezoelectric applications of polycrystalline κ -Ga₂O₃ thin films.

Author contributions

Yoshiaki Hirai: methodology, formal analysis, investigation, data curation, visualization, writing – original draft. Htet Su Wai: investigation. Toshiyuki Kawaharamura: resources, investigation. Noriaki Ikenaga: formal analysis, investigation, resources. Osamu Ueda: formal analysis, investigation. Hiroyuki Nishinaka: conceptualization, project administration, funding acquisition, supervision, resources, writing – review & draft.

Conflicts of interest

There are no conflicts to declare.

Data availability

Data will be made available upon request.

Acknowledgements

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