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Journal of Crystal Growth 278 (2005) 638–642
JOURNAL OF CRYSTAL GROWTH
www.elsevier.com/locate/jcrysgro

Thin single-crystal Sc_2O_3 films epitaxially grown on Si (1 1 1)—structure and electrical properties

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Abstract

Single-crystal single-domain Sc_2O_3 films have been epitaxially grown on Si (1 1 1) using molecular beam epitaxy (MBE) techniques. The Sc_2O_3 films have the bulk bixbyite cubic phase with a very uniform thickness, a structural perfection, and a sharp interface with Si. The thin oxide films exhibit bright, streaky, and reconstructed RHEED patterns. The high-intensity oscillation in the reflectivity, the strong Pendollusung fringes around the Sc_2O_3 (2 2 2) diffraction peak, and their narrow rocking curves are observed using the high-resolution X-ray diffraction (XRD). The (1 1 1) axis of the oxide films is parallel to the (1 1 1) axis of the Si substrate. The cone scans of the Sc_2O_3 {4 4 0} and Si {2 2 0} diffraction peaks about the surface normal find a 60° symmetry rotation of the film with the respect to the substrate. The Sc_2O_3 films exhibit low electrical leakage currents and a breakdown field of more than 5 MV/cm.

PACS: 68.55.−a; 61.10Nz; 61.10Kw; 61.14Hg

Keywords: A3.Epitaxially grown; A3.MBE; B1.Sc_2O_3

1. Introduction

Hetero-epitaxy between insulators and semiconductors has always attracted a lot of interest in many branches of science and technology. Typical examples are the growth of GaN and its related compounds on sapphire, which provides a basis for blue, green lasers, LEDs, and future lighting industry. Another example is found in the epitaxial growth of insulators on Si, which has potential applications in Si-on-insulator and high k gate dielectrics (to replace SiO_2) on Si. An epitaxial

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0022-0248/$ - see front matter
doi:10.1016/j.jcrysgro.2004.12.084

growth of other semiconductors such as GaN and GaAs on these insulating single-crystalline layers, thus indirectly on Si, may provide a basis for integrating photonic and high-power devices with the most advanced electronic Si devices. However, unlike homo-epitaxy such as GaAs epilayers grown on GaAs substrates, it is extremely difficult to achieve a structural perfection in the hetero-epitaxy, as witnessed in the growth of GaN on sapphire.

In this paper, we report an excellent epitaxial growth of single-domain, single-crystal Sc_2O_3 films on Si (1 1 1), an unexpected result. The structural perfection of the Sc_2O_3 films 18 nm thick is evidenced from very bright streaky and reconstructed RHEED patterns, very narrow rocking curves in the high-resolution X-ray diffraction (XRD), and a flat smooth film both at the interface and in the film interior observed using X-ray reflectivity and HR-transmission electron microscopy (TEM). Decent electrical characteristics of the oxide film were measured with a low leakage current and a high breakdown of >5 MV/cm. Previously, Sc_2O_3 was found to effectively passivate GaN [1] and to grow single crystal on GaN and sapphire (Al_2O_3), but with the in-plane growth in two degenerate orientations [2].

2. Experimental procedure

Si wafers with (1 1 1) as the normal to the wafer plane were put into a multi-chamber MBE/UHV system [3]. The Si wafers were neither RCA-cleaned nor with an HF dip before they were put in the HUV chamber. Therefore, the native oxides remained on top of the Si wafers. Nevertheless, heating the Si wafer to temperatures above ~890 °C with simultaneous deposition of a fraction of a monolayer of Si has resulted in a sharp streaky RHEED pattern, indicative of the removal of the native oxides. Additional growth of Si films ~3 nm thick ensures a chemically clean, atomically ordered, and (7×7) reconstructed Si(1 1 1) surface. The wafer was then transferred under UHV to an oxide chamber for the Sc_2O_3 deposition. Sc_2O_3 films were deposited by electron beam evaporation of pure powder-packed Sc_2O_3 source at substrate temperatures of 770 °C. During the oxide deposition, the vacuum in the chamber was maintained in the low 10^{-9} Torr. Streaky (4×4) film RHEED patterns along the major in-plane axes of Si were observed after the oxide growth of ~1 nm thick, indicative of an in-plane alignment between the oxide film and the Si substrate. After the oxide growth, an amorphous Si cap layer 2.4 nm thick was in situ deposited to protect the thin Sc_2O_3 film.

Additional and more accurate structural measurements were carried out using single-crystal XRD on a triple-axes four-circle diffractomer with a 2 kW Cu-K_α source. The oxide film 18 nm thick is very uniform over the 2-in wafer as studied from the X-ray reflectivity measurement and TEM. The TEM sample analytical work was performed using a Philips TECNAI-20 FEG type TEM. The capacitance-voltage (*C–V*) and current-voltage (*I–V*) characteristics were measured using Agilent 4284A and Agilent 4156C.

3. Results and discussion

The streaky oxide film RHEED patterns along the in-plane axes of [1 1 0] and [1 1 2] of Si were shown in Fig. 1. The epitaxial oxide film has shown the same in-plane symmetry as that of the Si (1 1 1) substrate. The brightness and the sharpness of the RHEED of the thin Sc_2O_3 film is perhaps among one of the best RHEED observed for any hetero-epitaxially grown oxide films.

The crystal structure of the Sc_2O_3 film is determined to be the cubic phase. Fig. 2 shows a single-crystal X-ray scan along the surface normal around Si (1 1 1). Aside from the strong peaks from the Si (1 1 1) substrate and the Sc_2O_3 (2 2 2), we notice striking fringes near 5000 arcsec, which belong to Sc_2O_3 (2 2 2) plane. These well-defined Pendellosung oscillations caused by dynamic XRD are very sensitive to the perfection of the atomic structure. Modest imperfections are known to completely destroy these fringes. The evidence strongly testifies the high quality of the epitaxial Sc_2O_3 film and also indicates that the film is very uniform with smooth surface and interface.

A strong intensity oscillation at small-angle reflectivity (see in Fig. 3) again indicates that the

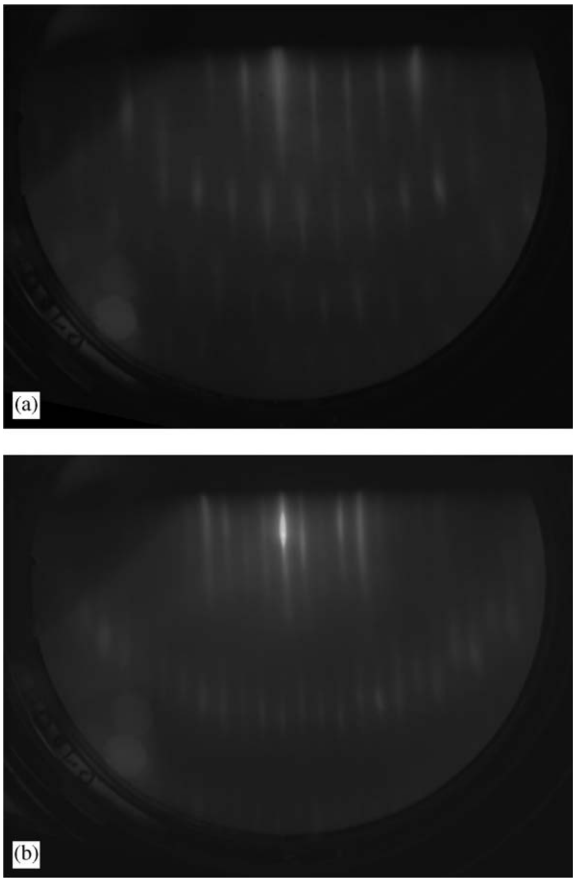


Fig. 1. In situ RHEED patterns of the Sc_2O_3 film 5 nm thick showing a (4×4) reconstruction showing (a) $[4\bar{2}\bar{2}]$ and (b) $[2\bar{2}0]$ directions.

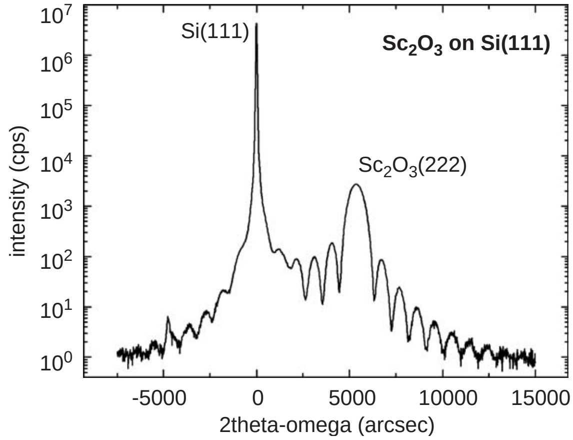
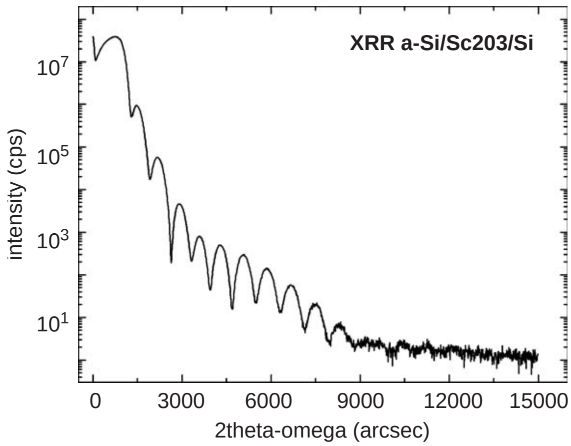


Fig. 2. Single crystal X-ray scan of the thin Sc_2O_3 film along the surface normal of the Si(1 1 1) substrate.



	material	thickness	roughness
Sub	Si		6.0Å
1	Sc_2O_3	181.2 Å	11.93 Å
2	a-Si	24.8 Å	11.48 Å

Fig. 3. Small-angle X-ray reflectivity measurement studying the roughness of the air/Si, Si/Sc_2O_3, and Sc_2O_3/substrate interfaces, and determining the film thickness to be 181 Å. (see Table of Fig. 3)

oxide film is highly uniform with a smooth Sc_2O_3/Si interface. Note that the intensity measurement here covers eight orders of magnitude. The small angle reflectivity quantifies a fairly accurate film thickness of ~181 Å. The analysis of the reflectivity measurement gives not only the oxide thickness but also the thickness of the Si cap layer, as listed in the table of Fig. 3. The interfacial roughness between Sc_2O_3/Si substrate, the Si cap/Sc_2O_3, and air/Si cap is estimated to be around 6, 11.93, 11.48 Å, respectively. The roughness has been improved dramatically in samples, in which the Si wafers were RCA cleaned and HF dipped prior to the oxide growth.

The FWHM of the rocking curves of Si (1 1 1) and Sc_2O_3 (2 2 2) are 10 (close to the XRD resolution) and 97 arcsec, respectively. The narrow rocking curve of the Sc_2O_3 (2 2 2) again indicates a high-quality oxide film. The ⟨1 1 1⟩ axes of the film and the Si substrate are well-aligned. All the other unit cell vectors of the films and the substrate

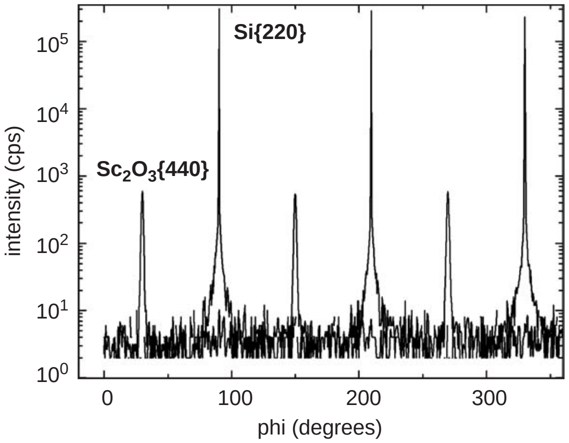


Fig. 4. Phi cone scans of Si{220} and Sc_2O_3{440}.

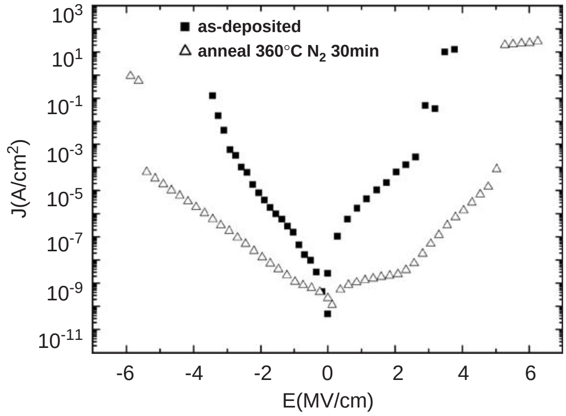


Fig. 5. Dependence of leakage current density on electric field for Sc_2O_3 film 181 Å with an undoped amorphous Si cap layer 24.8 Å.

are parallel, as studied using the pole-figure scans of Sc_2O_3 {4 4 0} peaks. The in-plane cone scans of the Sc_2O_3 {4 4 0} and Si {2 2 0} diffraction peaks about the surface normal find a 60° in-plane symmetry rotation of the film with the respect to the substrate surface orientation (Fig. 4). In contrast to a 6-fold symmetry found in the growth of Sc_2O_3 on sapphire or GaN [2], the 3-fold symmetry indicates the attainment of a single domain of the oxide films in the present work. These cone scans are made by tilting the χ-angle of the oxide film to bring the (4 4 0) reflection into the scattering plane, and then by scanning the π-angle over a 360° rotation. A pole figure around $\langle 1\,1\,1 \rangle$ axis of a cubic crystal should have three {4 4 0} peaks, as obtained in the Sc_2O_3 film.

From the TEM picture (not shown), there was no silicon oxide formed between the Si substrate and the Sc_2O_3 oxide film [4]. This indicated that the approach with e-beam evaporation is excellent in keeping the Sc_2O_3/Si interface clean and sharp. Also the smooth interface revealed using TEM is consistent with the studies using RHEED and X-ray reflectivity. Plan-view and cross-sectional TEM observations revealed that the crystal quality of the thin oxide film was almost defect-free with distinct Morie fringes and strain contrast: the oxide film was highly strained in order to accommodate the existing lattice mismatch. However, no structural defects, such as threading dislocations and micro-twins, were found morphologically.

Fig. 5 shows *J* vs. *E* characteristics for MOS diodes made of Sc_2O_3 films on Si with Au as a top electrode. Here *J* is the leakage current density as the leakage current divided by the measured area of $7.85 \times 10^{-5}\,\mathrm{cm}^2$, and *E* is the electrical field as the biasing voltage divided by the thickness of the oxide and the amorphous Si cap layer, which may have been partially oxidized when removed outside UHV. The positive bias means that the top metal electrode is positive with respect to Si. Based on *J*–*E* results on the samples of as-deposited condition and annealing at 360 °C under a flow of pure N_2 for 30 min, it seems to suggest that the annealing reduces the defects in the film, which may be produced by the secondary electron bombardment during the film growth. The leakage current was drastically reduced, for example from $10^{-3}\,\mathrm{A/cm^2}$ to less than $10^{-7}\,\mathrm{A/cm^2}$ at 2 MV/cm. The breakdown behavior of the oxide is symmetric and breakdown field has been improved to >5 MV/cm.

4. Conclusions

To our knowledge, the crystalline quality of Sc_2O_3 on Si (1 1 1) is the best among all the single-crystal oxides hetero-epitaxially grown on foreign substrates. The bulk lattice constants of Si (5.43 Å) and Sc_2O_3 (9.86 Å) are mismatched by 9.2%, if we take the double Si unit cell dimension. It is, therefore, amazing that the Sc_2O_3 grows very well epitaxially on Si. The in-plane symmetry of the Sc_2O_3 is similar to those of Gd_2O_3 and Y_2O_3 epitaxially grown on GaN and sapphire [5]. Single-crystal GaN was grown epitaxially on both of the rare earth oxide thin films [6] without growth of any intermediate layers as in the growth of GaN on sapphire substrates. It is anticipated that single-crystal GaN and its related compounds will be grown epitaxially on the Sc_2O_3, thus indirectly on Si. The structural perfection in the Sc_2O_3 single-crystal also translates into the measured low electrical leakage currents and a high breakdown field, indicating that not too many electrical-induced defects are generated due to the large lattice mismatch between the oxide films and the Si substrate.

Acknowledgments

We would like to express our thanks for the supports from the National Science Council under contract NSC92-2119-M-007-054 and the Center for Nano Science and Technology of University System of Taiwan, Republic of China.

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