Optical properties of gallium oxide thin films

M. Rebien(a) and W. Henrion
Hahn-Meitner-Institut, Abteilung Silizium-Photovoltaik, Kekuléstr. 5, D-12489 Berlin, Germany

M. Hong and J. P. Mannaerts
Agere Systems, Electronic Devices Research Laboratory, Murray Hill, New Jersey 07974

M. Fleischer
Siemens AG, Corporate Technology, Otto-Hahn-Ring 6, D-81739 Munich, Germany

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The stable oxide of gallium, monoclinic $\beta$-Ga$_2$O$_3$, is a wide band gap material. While current applications include luminescent phosphors and gas sensors it has been recognized as a promising candidate for deep-ultraviolet transparent conductive oxides (deep-UV TCO). In addition, $\beta$-Ga$_2$O$_3$ may be applied in textured dielectric coatings for solar cells. A value of the refractive index close to $\sqrt{n_{GaAs}}$ allows the preparation of efficient single-layer antireflective coatings for GaAs. Renewed interest with respect to the passivation of GaAs surfaces arose in connection with the realization of a very low electronic interface state density using $\beta$-Ga$_2$O$_3$ single crystals. X-ray diffraction revealed a nanocrystalline morphology with a crystallite size of $\approx$10 nm.

Accurate determination of the optical functions is an essential prerequisite for device simulations and gives the opportunity to improve material preparation. The anisotropic absorption edge of $\beta$-Ga$_2$O$_3$ single crystals has been investigated thoroughly. However, refractive index dispersion of high-quality material in the uv–visible–near infrared (UV–VIS–NIR) spectral range has not been reported. Published refractive index data are either restricted to a single spectral position or have been determined on material with a high density of structural defects.

Here we report the linear optical properties of electron-beam deposited and sputtered $\beta$-Ga$_2$O$_3$ thin films. A multisample analysis of ellipsometric spectra recorded at multiple angles of incidence was performed to extract the optical functions using appropriate layer models. A comparison is made between refractive index spectra of both types of films and the absorption edges are determined.

$\beta$-Ga$_2$O$_3$ films on GaAs were prepared by electron-beam evaporation of $\beta$-Ga$_2$O$_3$ pellets at a growth rate of about 0.5 Å/s. According to transmission electron microscopy investigations the films consist of randomly distributed microcrystallites within an amorphous matrix. The films were deposited onto an epitaxial GaAs buffer layer (Si doping density $1.6 \times 10^{16}$ cm$^{-3}$) on (001)-oriented GaAs substrate wafers (Si doping density $2 \times 10^{18}$ cm$^{-3}$).

The optical functions of $\beta$-Ga$_2$O$_3$ thin films have been determined by ellipsometry from 0.74–5 eV. Several electron-beam evaporated and rf magnetron sputtered films of different thicknesses were investigated using a multisample technique. Refractive index values comparable to those of bulk material are found. Cauchy dispersion model fits yield a high-frequency dielectric constant $\varepsilon_\infty$ of 3.57. Above 4.7 eV a direct absorption edge is observed. © 2002 American Institute of Physics. [DOI: 10.1063/1.1491613]
range of 0°–360° and of partial depolarization. For comparison, near-normal hemispherical reflectance (~) native SiO2 interlayer was included in the model. Surface layer on a semi-infinite substrate. For films on silicon the fore, we used the dataset of Zollner for intrinsic GaAs. 21 datasets for doped GaAs are not sufficiently accurate. There-

tivity and on accuracy of the substrate optical functions. The model based analysis critically depends both on model qual-

tion weigthed to the experimental error allows to extract the 

FIG. 2. Refractive index spectra derived from the multisample fits. Cauchy dispersion model parameters are given in Table II. Refractive index ranges from literature are included for electron-beam deposited films (see Ref. 11), as sprayed and annealed thin films (see Ref. 12) as well as bulk material (see Ref. 13).

FIG. 3. Imaginary part of the dielectric function \( \varepsilon_2 = 2nk \) in the plot for direct allowed transitions. Linear extrapolation to determine the direct band gap is indicated by dashed lines.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Deposition method</th>
<th>Substrate</th>
<th>Film thickness (nm)</th>
<th>Max thickness inhomogeneity</th>
<th>Surface roughness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Electron-beam evaporation</td>
<td>GaAs</td>
<td>31.9±0.1</td>
<td>9%</td>
<td>2.8±0.1</td>
</tr>
<tr>
<td>2</td>
<td>Electron-beam evaporation</td>
<td>GaAs</td>
<td>58.95±0.07</td>
<td>4.8%</td>
<td>3.8±0.1</td>
</tr>
<tr>
<td>3</td>
<td>Sputtering</td>
<td>Si</td>
<td>127.7±0.2</td>
<td>3%</td>
<td>…</td>
</tr>
<tr>
<td>4</td>
<td>Sputtering</td>
<td>Si</td>
<td>597.5±0.2</td>
<td>0.9%</td>
<td>…</td>
</tr>
<tr>
<td>5</td>
<td>Sputtering</td>
<td>Si</td>
<td>2468±0.6</td>
<td>0.45%</td>
<td>…</td>
</tr>
</tbody>
</table>

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film thicknesses. Fit parameter correlation was found to be generally small. Only the surface roughness of the sputtered films was strongly correlated to the thickness of the interfacial SiO₂ (Table I). Depolarization spectra (not shown) were fitted equally well. The rough sample backside suppressed specular incoherent reflections leaving film thickness non-homogeneity and spectral bandwidth as possible causes of sizeable depolarization for the highly specularly reflecting films under investigation. The influence of the spectral bandwidth set by the monochromator was small compared to film thickness variations. Depending on the measurement spot size, which is a function of the angle of incidence, different values of depolarization have been observed. Hence, the thickness non-homogeneity values were fitted separately for each angle of incidence. The maximum values are given in Table I. The use of more complicated layer models leads to strong fit parameter correlations and does not significantly improve the fit. Further confirmation for the modelling is indicated by a good agreement between measured reflectance and those calculated using the film optical functions determined ellipsometrically.

In Fig. 2 the resulting refractive index spectra are shown together with comprehensive values from literature. Even for the two very different preparation methods and substrates as well as the wide range of film thickness of the investigated samples, the resulting dispersion curves are nearly identical. As a maximum error for the β-Ga₂O₃ refractive index the maximum difference between both curves of 0.02 near 3.8 eV can be taken, although the measurement and modelling errors are smaller. This deviation is attributed to small variations between different deposition methods and runs, differences in film morphology, or small inhomogeneity within the films. A high-frequency dielectric constant \( \epsilon_{\infty} \) of 3.57 (corresponding to \( n_\infty = 1.89 \)) was deduced from the Cauchy model fits (Table II). Absolute \( n \) values in the range of bulk material indicate a compact nature of the films. No clear signs of refractive index anisotropy were observed. For the determination of the anisotropic optical functions of this monoclinic material the investigation of high-quality single crystals is required.

Absorption in the films is below the detection level in most of the spectral range investigated. The direct absorption edge (Ref. 27) was obtained from the imaginary part of the dielectric function \( \epsilon_2 = 2nk \) (Fig. 3). The values of 4.72 eV (sputtered β-Ga₂O₃) and 4.74 eV (e-beam evaporated β-Ga₂O₃) are close to that of single crystals with E∥b light polarization. Small absorption below this value and a likely lower edge around 4.5 eV as reported in Refs. 1 and 14 cannot unambiguously be distinguished from layer model inaccuracies. The comparatively larger absorption of the electron-beam evaporated material above 4.5 eV compared to the sputtered films is attributed to inaccurate representa-

<table>
<thead>
<tr>
<th>Deposition method</th>
<th>spectral range (µm)</th>
<th>( n_\infty )</th>
<th>B (µm(^2))</th>
<th>C (µm(^4))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron-beam evaporation</td>
<td>0.295–0.826 (1.5–4.2 eV)</td>
<td>1.891±0.002</td>
<td>0.0110±0.0005</td>
<td>0.00048±0.00004</td>
</tr>
<tr>
<td>Sputtering</td>
<td>0.342–1.630 (0.76–3.62 eV)</td>
<td>1.883±0.0003</td>
<td>0.0114±0.00007</td>
<td>0.000359±0.000007</td>
</tr>
</tbody>
</table>

18 R. M. A. Azzam and N. M. Bashara, Ellipsometry and Polarized Light (Elsevier, Amsterdam, 1987).