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Optical properties of nanocrystalline Y_2O_3 thin films grown on quartz substrates by electron beam deposition

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ABSTRACT

Yttrium oxide thin films of a thickness 221–341 nm were formed onto quartz substrates by reactive physical vapor deposition in an oxygen atmosphere. An electron beam gun was applied as a deposition source. The effect of substrate temperature during film deposition (in the range of 323–673 K) on film structure, surface morphology and optical properties was investigated. The surface morphology studies (with atomic force microscopy and diffuse spectra reflectivity) show that the film surface was relatively smooth with RMS surface roughness in the range of 1.7–3.8 nm. XRD analysis has revealed that all diffraction lines belong to a cubic Y_2O_3 structure. The films consisted of small nanocrystals. Their average grain size increases from 1.6 nm to 22 nm, with substrate temperature rising from 323 K to 673 K. Optical examinations of transmittance and reflectance were performed in the spectral range of 0.2–2.5 μm. Optical constants and their dispersion curves were determined. Values of the refractive index of the films were in the range of n = 1.79–1.90 (at 0.55 μm) for substrate temperature during film deposition of 323–673 K. The changes in the refractive index upon substrate temperature correspond very well with the increase in the nanocrystals grain diameter and with film porosity.

1. Introduction

Last year's thin films of yttrium oxide (Y_2O_3) have been the subject of intensive examinations, due to very interesting physicochemical properties [1–29]. At normal pressure yttrium oxide exhibits a cubic C-type structure of rare-earth oxide (bixbyite, Mn_2O_3-type structure) with the lattice constant of 1.06 nm [9,23]. The lattice constant of Y_2O_3 matches very well the lattice constant of Si (being approximately two times the Si lattice parameter). It makes it a very useful material for silicon-based nanotechnology. Yttrium oxide is characterized by a wide band gape (E_g = 5.91–6.15 eV) [9], high optical transparency over a broad wavelength range (0.2–8 μm) [3,8,9,25], low absorption (from near-ultraviolet (UV) to infrared (IR) [9]) having the refractive index in the range of 1.7–1.93 at 0.55 μm [4,8,9,21,25,28,29]. Y_2O_3 exhibits also high thermal conductivity: 0.13 W cm^{-1} K^{-1} [8]. Thin films of Y_2O_3 examined in metal–insulator–metal (MIM), or metal–insulator–semiconductor (MIS) structures are characterized by high dielectric permittivity (ε = 5.1–31.6) [1,3,10,20,22,25–27] and superior electrical breakdown field strength (F_b = 7–10.7 MV/cm) [2,4,6,15,20]. Taking into account all mentioned properties, yttrium oxide seems to be an excellent and promising material for fabrication of various optical, electronic and optoelectronic thin film devices, such as MIM capacitors [5], MIS structures [3,5,6,10], planar waveguides [8,12] and interference coatings [24].

Several physical vapor deposition (PVD) techniques have been applied for fabrication of Y_2O_3 thin film coatings and Y_2O_3-based film structures, namely: radio-frequency (RF) magnetron sputtering [4–11,28], pulsed laser deposition [13–15], electron beam (EB) deposition [16–24], ion beam assisted deposition (IBAD) [25,26], molecular beam epitaxy [27] and R-F plasma assisted metal organic chemical vapor deposition (MOCVD) [29]. In this paper we have fabricated Y_2O_3 films with the use of the EB deposition method. All deposition processes were carried out as a reactive deposition of yttrium oxide in an oxygen atmosphere. In the paper [30] we have described dielectric properties of Y_2O_3 films. The aim of this report is to present basic optical characteristics of films deposited at various substrate temperatures and to correlate their optical properties with the film microstructure.

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2. Experimental methods

2.1. Sample preparation

All thin-film dielectric coatings were formed by physical vapor deposition of yttrium oxide onto quartz plates at various substrate temperatures, from 323 K to 673 K. Before the deposition vacuum apparatus was pumped down to about 10^{-7} Torr (1.3×10^{-5} Pa). The film depositions were carried out at the oxygen pressure of 8×10^{-5} Tr (1×10^{-2} Pa) as the reactive deposition process. A water-cooled, electron gun of the power of 10 kW was used as an evaporation source. Y_2O_2 of 99.9% purity, fabricated by Koch-Light-Laboratories, was used as a source material. Film growth (thickness and deposition rate) was monitored during the evaporation process with a crystal (quartz) monitor attached to the vacuum system. All film deposition parameters for specimens denoted as A, B, C and D are given in Table 1.

2.2. Measurement procedures

Film thickness measurements were performed with the use of the multiple-beam interference method (the Tolansky's method). An optical interference microscope working at the wavelength of 546 nm was applied to this aim. Y_2O_2 films fabricated by us had a thickness in the range of 221–341 nm.

The X-ray Diffraction (XRD) measurements were carried out by a Philips diffractometer supported by a parallel beam optic and Cu Kα_1 radiation source (at the wavelength λ = 1.540597 Å). The grain size of nanocrystals was estimated from the well-known Scherrer equation:

$$D = \frac{k \cdot \lambda}{\beta \cdot \cos\theta} \tag{1}$$

where β is the full width at half maximum of the diffraction peak, θ is the Bragg angle and k is the shape factor ($k = 0.9$).

Atomic force microscopy (AFM) measurements and surface topography observations were made with XE-70 PARK-System. The data were taken in ambient air in a noncontact mode.

Optical properties of Y_2O_2 thin film coatings were examined in the range of wavelengths from 0.2 µm to 2.5 µm. The Jasco type V-570 two-beam spectrophotometer was used for transmittance and reflectance measurements. For detection of diffuse reflectivity spectra, the spectrophotometer was equipped with an integrating sphere. These measurements were performed in the wavelength range of 196–800 nm.

The transmittance measurements (T_m) of the specimens were carried out with relation to the transmittance of the same quartz plate without any coating ($T_{m/q}$) and with relation to the air ($T_{m/a}$). Then, the transmittance (T) of the film (in the system: film-finite substrate) was evaluated as the average value obtained from Eq. (2a) or (2b), respectively [31]. The reflectance R_m of the sample was measured with respect to the standard mirror. The reflectance of the film was determined from Eq. (3).

$$T = \frac{T_{m/q}\left[1 - R_s R'\right]}{1 + R_s} \tag{2a}$$

$$T = \frac{T_{m/a}\left[1 - R_s \cdot R'\right]}{1 - R_s} \tag{2b}$$

$$R = R_m - \frac{T^2 R_s}{1 - R' \cdot R_s} \tag{3}$$

in which:

$$R' = \frac{R'_m - R_s}{\left[(1 - R_s)^2 + R_s(R'_m - R_s)\right]} \tag{4}$$

R_s is the reflectance of the quartz plate. In above equations, R_m (R) denotes the front measured (evaluated) reflectance for the film facing the incident light and the back measured (evaluated) reflectance R'_m (R') for a substrate facing the incident light, respectively.

Table 1
Deposition parameters of yttrium oxide thin films.

Y_2O_3 sample no.	Film thickness (nm)	Substrate temperature (T_s) (K)	Deposition rate (nm/s)
A	235.2 ± 1.4	323	0.21
B	221 ± 2	390	0.18
C	385.8 ± 2	623	0.13
D	340.9 ± 0.6	673	0.1

3. Film characterization

3.1. X-ray diffraction

Fig. 1 shows the XRD spectra of yttrium oxide films deposited under technological conditions specified in Table 1. The diffraction patterns show that for all samples a few diffraction lines superimposed on an amorphous response of the quartz substrate are observed. All the diffraction patterns correspond to the C-type cubic phase of Y_2O_3 (identified according to JCPDS card 25-1200). The films deposited at substrate temperature of 323 K and 390 K were almost amorphous. Only two very broad and weak reflections corresponding to (431) and (662) planes were observed on the XRD patterns. Low intensity of these reflections is connected with very small (nanometric) size of grains. Internal stress in the film can have also influence on the position of these peaks. For films deposited at higher substrate temperature, the additional lines of (222), (400), (332), (440), (622) planes appeared. The grain sizes were estimated using the Scherrer expression (Eq. (1)). The average grain sizes estimated in this way were in the range from 1.6 nm to

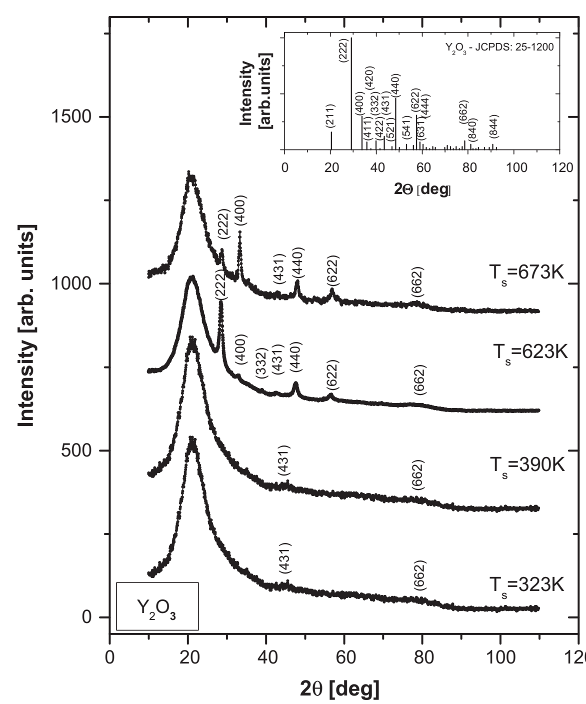

Fig. 1. XRD pattern of yttrium oxide films deposited onto quartz plates at different substrate temperatures (T_s). The insert figure shows the reference data for cubic structure of Y_2O_3 (according to JCPDS card 25-1200).