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Comparison in formation, optical properties and applicability of DC magnetron and RF sputtered aluminum oxide films

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Amorphous aluminum oxide (a-Al_2O_3, alumina) can be widely used for ceramic coatings, gate oxide for microelectronics and waveguiding component of integrated optical elements. Moreover, it is a candidate for masks and molds for the preparation of new generation nanoscale devices. Among different technological procedures, cathode sputtering is one of the most effective techniques to deposit amorphous materials which could not be vitrified by an ordinary melting method. Here, the structural and optical properties of Direct Current (DC) magnetron and Radio Frequency (RF) sputtered alumina layers have been revealed regarding to the preparation method. It is shown that the optical absorption and the refractive index of the RF sputtered alumina enable the films to be used as high quality waveguiding material. The oxygen incorporation from the plasma with higher oxygen content results in a bandgap shift to the lower values. Contrarily, reactive DC magnetron sputtering process led to only partly oxidized film growth exhibiting higher absorption.

1. Introduction

The boom of telecommunication, data storage- and display applications, etc in the last decade affected micro- and optoelectronics industries to become one of the strongest drivers of market economy. The fast development of these technologies had expanded coating uses in diverse areas, as optical [1,2], protective, insulating or decorative thin films [3]. Thus, thin films deposition plays a key role in the development of the semiconductors, microelectronics, optoelectronics, optics and many other edge technologies [4]. Industry continuously demands films of improved quality and sophistication. The rapid development of deposition technologies is also due to the improved understanding of the physics and chemistry of films, surfaces, interfaces and microstructural mechanisms and it became possible to develop the periodic structures on the interfaces for different purposes. By way of example, an appropriate patterning of the surface, the basis of advanced functionality, is generally obtained by lithographic processes resulting in alternating film deposition, lithography and etching steps [5,6]. Simple patterning process has not been applied in industry yet. Self-organized patterning of supported nanoclusters is considered as one of the promising methods; however, this approach is puzzling, since the capability of controlling the patterns based on a suitable combination of clusters and templates [7]. demonstrate that Co nanoclusters grown from vapour deposition over Al_2O_3 thin films on NiAl(100) substrate make an appropriate combination for self-organized patterning.

The essential technological step, the metal oxides and oxide thin films deposition can be carried out by a large variety of technologies, although none of them allows easy in situ patterning. Aware of that, patterning is not possible for most of the deposition technologies; we still mention here a possible two step method using UV laser. This work was initiated from a question brought up by our research on nanostructuring of macroscopic area of the alumina films. We approached this question by adopting methods common in thin film technology. Alumina/aluminum/off-stoichiometric thin films were deposited by the reactive DC sputtering. Additional considerations were that thin films for optical waveguiding elements must be characterized by low propagation loss, by thickness and refractive index easily reproducible over a large substrate area. As a consequence, it is fundamental to prepare such composite systems by means of synthesis techniques in which the parameters affecting the optical properties could be varied in a controlled way.

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Sputtering technology is a promising solution for the fabrication of transparent, high refractive index, amorphous or crystalline oxide materials, which can satisfy the demands for next generation integrated photonics [8,9]. Alumina-stoichiometric thin films were deposited by RF sputtering. It is known that the refractive index of the a-Al_2O_3 film produced by RF sputtering from an Al_2O_3 target is markedly dependent on the deposition conditions: low index films tend to have higher sputter rates than high index films, which suggest that the low indices are due to film porosity [10]. Variable stoichiometry seems to be a better method to achieve controllable refractive index. For the sake of simplicity, we used alumina ceramic as a target and different plasma composition deposition processes.

2. Experimental details

2.1. Sample preparation

Thin aluminum oxide layers were deposited by RF and DC magnetron sputtering at room temperature onto Si (100) substrates. RF sputter deposition was carried out in a Leybold Z400 apparatus evacuated to $5*10^{-5}$ Pa. Sputtering was performed under a mixture of high purity argon and oxygen gases with an applied RF power of 255 W yielding a plasma pressure of $2.5*10^{-2}$ mbar. The target with diameter of 75 mm was coupled to the RF generator operating at 13.56 MHz. Oxygen was incorporated into the layers by flowing it continuously into the sputtering chamber at different flow rates, resulting partial pressure of oxygen up to 15% of the total one. We experienced that high purity ceramic (DUROX AL with a chemical purity of 99.8%) is a dense, non-porous, vacuum tight material that has a consistent dielectric constant which makes it excellent for sputtering targets. This ceramic (DUROX AL) target with diameter of 75 mm was coupled to the RF generator operating at 13.56 MHz. The disadvantage of the method mentioned above is that the deposition rate is as low as 2.4 nm/min.

DC magnetron sputtering is a higher deposition rate process than RF, however only electrically conductive materials can be used as targets. Therefore, an Al target (Aldrich, 99.999%, diameter of 50 mm) was applied for the reactive DC sputtering. The base pressure was $3*10^{-5}$ mbar, while the sputtering gas was a mixture of O_2-Ar with partial pressures of $4*10^{-4}$ mbar and $5*10^{-3}$ mbar, respectively. The power of the DC magnetron was adjusted to 55 ± 5 W.

2.2. Sample treatment and characterization

In the case of alumina prepared in the form of thin films, we investigated the relationships between deposition techniques and structural characteristics, optical parameters. The structure and morphology of the films were characterized by means of Transmission Electron Microscopy (TEM) and Selected Area Electron Diffraction (SAED). The TEM and SAED studies were carried out by a 200 kV Philips CM 20 and a 300 kV Jeol 3010 high resolution TEM.

Optical properties of thin film structures can be derived from spectroscopic ellipsometry (SE) measurement, which is known to be a high-precision optical characterization technique. SE determines the change in polarized light upon light reflection on a sample. SE measures two quantities, Ψ and Δ. These are the amplitude ratio Ψ and phase difference Δ between light waves known as p- and s-polarized light waves. The method of Variable Angle Spectroscopic Ellipsometry (VASE) makes possible measurements at multiple angles of incidence. An additional angle will change the length of the light penetrating through the materials. Multiple angles are helpful to improve the confidence limits of the results yielded by the evaluation of the measured spectra. The VASE measurements were performed with angles of incidence of 55°, 60°, 65° and 70°. The measured data were analyzed using the computer code of WVASE32 [10].

3. Results

3.1. Structure and morphology of the RF and DC sputtered alumina layers

It was found that the two films deposited by RF and DC magnetron sputtering show different morphologies. The cross-sectional TEM micrographs of a typical RF and a DC sputter-deposited alumina films are shown in Fig. 1. The RF sputtered film (A) appears as homogeneously amorphous, while the DC sputter deposited one (B) exhibits remarkable inhomogeneities that we attributed to embedded nanocrystals. The high resolution micrograph in Fig. 2(A) represents a completely amorphous film that is indicated, also, by the diffuse rings in the SAED inset. Contrary to that, the DC sputter-deposited layer in Fig. 2(B) shows individual Al nanocrystals of a few nanometer size embedded in the amorphous Al-oxide matrix. The arrows mark such nanocrystals with resolved lattice planes. This is supported by the SAED inset in (B), that, in addition to the diffuse rings, shows relatively sharp ones that can be indexed as fcc Al. It is to mention that the SAED patterns are taken from identical plan-view samples (not shown here) by means of a selected area aperture of 200 μm sizes. This allows the selection of a sample area of ~5 μm in diameter.

3.2. Results of evaluation of SE data

The optical model applied in the evaluation of the measured SE data consisted of two layers on top of single crystalline silicon. The layer adjacent to the single crystalline silicon substrate represents the sputtered aluminum oxide layer and the surface roughness was considered as the second layer. The roughness layer was taken into account on basis of effective medium approximation; the roughness layer consists of 50% of the sputtered aluminum oxide and 50% of void.

Tauc and his coworkers suggested an expression for the imaginary part of the refractive index above the band edge [11]. Jellison and his coworkers elaborated the so called Tauc-Lorentz model that gives an expression for the imaginary part of the dielectric function if only a single transition is considered [12,13]. One obtains the Tauc-Lorentz model by multiplying the Tauc expression for the imaginary part of the dielectric function near the band edge by the imaginary part of the complex dielectric function of a single Lorentz oscillator:

$$\begin{aligned} \varepsilon_2 &= \frac{AE_0\Gamma(E-E_g)^2}{\left(E^2-E_0^2\right)^2+\Gamma^2E^2}\frac{1}{E}, \quad E>E_g \\ \varepsilon_2 &= 0, \quad E \le E_g \end{aligned} \tag{1}$$

The model contains the following parameters: band gap E_g; peak transition energy E_0; broadening parameter Γ; and a factor A, which represents the optical transition matrix elements. E represents the photon energy.

The real part of the dielectric function in the Tauc-Lorentz model is obtained by the Kramers-Kronig integration. The Tauc-Lorentz model is consistent with fundamental considerations and appears to provide a good representation of dielectric functions for disordered materials with only five parameters, although the expression is complicated [12–15].

The Cauchy dispersion model is given by the equation: