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# Optical scattering characteristic of annealed niobium oxide films

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**Abstract**

Niobium oxide ($Nb_2O_5$) films with thicknesses ranging from 200 to 1600 nm were deposited on fused silica at room temperature by low frequency reactive magnetron sputtering system. In order to study the optical losses resulting from the microstructures, the films with 500 nm thickness were annealed at temperatures between 600 and 1100 °C, and films with thicknesses from 200 to 1600 nm were annealed at 800 °C. Scanning electron microscopy and atomic force microscopy images show that the root mean square of surface roughness, the grain size, voids, microcracks, and grain boundaries increase with increasing both the annealing temperature and the thickness. Correspondingly, the optical transmittance and reflectance decrease, and the optical loss increases. The mechanisms of the optical losses are discussed. The results suggest that defects in the volume and the surface roughness should be the major source for the optical losses of the annealed films by causing pronounced scattering. For samples with a determined thickness, there is a critical annealing temperature, above which the surface scattering contributes to the major optical losses. In the experimental scope, for the films annealed at temperatures below 900 °C, the major optical losses resulted from volume scattering. However, surface roughness was the major source for the optical losses when the 500-nm films were annealed at temperatures above 900 °C.



*Keywords:* Optical coatings; Niobium oxide; Optical properties; Surface roughness

## 1. Introduction

Since amorphous $Nb_2O_5$ films have unique physical and chemical properties, such as high refractive index, excellent chemical stability and corrosion resistance, and very low optical absorption in the visible and near infrared light range, they are widely used as high quality optical interference filters [1,2]. However, in general, $Nb_2O_5$ is a complex material and can show a great variety of polymorphic forms and various structures [3]. For magnetron sputtered films, because of the various energetic processes, such as elevated substrate temperature, ion or electron bombardment, crystal growth or re-crystallization may take place. This will in turn result in optical losses due to the scattering of light by the grains and grain boundaries. Moreover, the surface roughness, microcracks and voids in the films which come from the deposition process would also induce optical losses [4–6]. Clarifying the correlation between the microstructure and the optical loss is an important issue for fabricating high quality optical films.

Tremendous efforts have been devoted to the understanding of the influences of microstructure on the optical properties of $Nb_2O_5$ films [1,2,7–9]. For instance, Venkataraj et al. have studied the annealing effects on the structural and optical properties of hexagonal structure (TT phase) $Nb_2O_5$ films at temperatures between 400 and 600 °C [7], and Lee et al. have reported the influence of ion-beam energy on the internal stress and optical properties of niobium oxide films [2]. It is also reported that the optical properties of $TiO_2$ films annealed at high temperatures can

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be drastically affected by the structural and morphological changes [10,11].

In this paper, we focus on the influences of the surface roughness and defects, such as voids, microcracks, grains and grain boundaries, on the optical losses of $Nb_2O_5$ films deposited on fused silica substrates by reactive magnetron sputtering. The surface roughness and defects changed during post-annealing of the as-deposited samples with different film thicknesses. The results show that the root mean square (RMS) of surface roughness of 500-nm films increased from 1.3 nm for the as-deposited films to 67 nm for the film annealed at 1100 °C. Meanwhile, microcracks, voids and grain size increased drastically after annealing at high temperatures. For the films annealed at 800 °C with different thicknesses, RMS surface roughness, voids and grain size also increased as thickness increased. Correspondingly, the optical transmittance decreased and the optical loss increased as the annealing temperature or thickness of films increased. The optical scattering resulted from both surface roughness and defects in volume were discussed. The results showed that the surface roughness was the major source for optical scattering when 500-nm films were annealed at temperatures above 900 °C. However, when the films were annealed below 900 °C, volume scattering became the major source of scattering.

## 2. Experimental details

Niobium oxide thin films were prepared on unheated fused silica substrates by using a low frequency reactive magnetron sputtering system (ASC-800, Shincron Co. Ltd.). Pure (99.9%) niobium target, cooled with water, was located 12 cm away from the substrate. The power of sputtering was kept at 3.98 kW. The base pressure of the vacuum chamber was $2.0\times10^{-4}$ Pa and the pressure was kept at $1.67\times10^{-1}$ Pa with argon/oxygen mixed gas during the sample preparation. The flow rates of argon and oxygen were individually kept at 100 sccm using two mass-flow controllers. Prior to depositing $Nb_2O_5$ films, the targets were presputtered for 5 min to remove their surface oxides. The annealing process of the films was performed in an oven between 600 and 1100 °C for 1 h in air.

The crystal structure and phase of the films were examined by X-ray diffraction (XRD). XRD study was carried out on an 18 kW MXPAHF X-ray diffractometer with high-intensity $CuK_\alpha$ radiation ($\lambda$ = 1.5418 Å).

The cross-sectional microstructure and surface of the films were observed by a field-emission scanning electron microscopy (SEM) (JSM-6700F). The morphology of $Nb_2O_5$ films was also observed with an atomic force microscopy (AFM) (SPA300HV) under ambient conditions. The RMS surface roughness of the films was calculated from AFM images. Scans were taken under a contact force of $10^{-9}$ N and over areas of $10\times10$ μm$^2$ for RMS surface roughness calculation. The reflectance and transmittance, from which the refractive index and absorption coefficients can be obtained, were measured in the $250<\lambda<1000$ nm wavelength range by a spectrophotometer (U-4100, Hitachi Co.) equipped with an integrating sphere. The reflectance and transmittance measurements were performed at near normal incidence (5°). The optical path (24 cm) from the sample to the integrating sphere is much larger than the aperture size (1 cm) of the integrating sphere. Therefore, the forward scattering component was negligible in our optical scattering loss experiment. The thicknesses of films were calculated from optical transmittance and were confirmed by the cross-sectional images observed by SEM.

## 3. Results and discussion

### *3.1. Crystal structure*

Fig. 1 shows the XRD results of niobium oxide films annealed for 1 h at various temperatures and thicknesses. As shown, the as-deposited films are amorphous and change into TT phase after annealing at 600 °C (Fig. 1a,b), which is consistent with the previous reports [3,7]. When the temperature increases, primitive orthorhombic structure (T phase) occurs in the $Nb_2O_5$ films at annealing temperatures from 700 to 1000 °C (Fig. 1c), while monoclinic end-centered structure (H phase) occurs at 1100 °C. The films annealed at 800 °C with different thicknesses are also T phase (Fig. 1d). The lattice parameters were calculated from the peak positions, and the corresponding structures can be identified by comparing our results with data from JCPDS international diffraction data base, as summarized in Table 1.

### *3.2. Microstructure and surface morphology*

As shown in Fig. 2a, no column structures or defects can be observed in the as-deposited films. Fig. 2b–e indicate the surface morphology and cross-sectional SEM images of $Nb_2O_5$ films at different annealing temperatures and thicknesses. For the 500-nm films, surface roughness

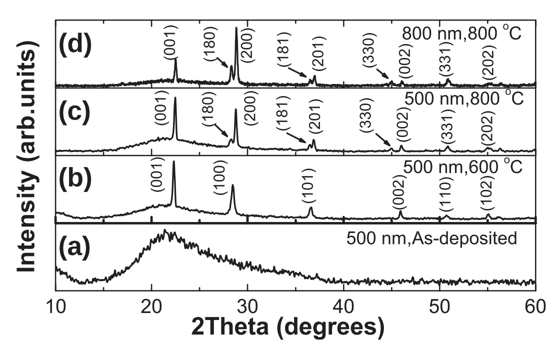


Fig. 1. X-ray diffraction patterns for niobium oxide films on fused silica substrates: (a) 500-nm film, as-deposited; (b) 500-nm film, annealed at 600 °C; (c) 500-nm film, annealed at 800 °C, and (d) 800-nm film, annealed at 800 °C.