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A study on sputtered ultrathin chromium films for optical applications

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ABSTRACT

Structural, optical, and electrical properties of ultrathin chromium films manufactured using magnetron sputtering were investigated. The films showed pure metallic chromium phase yet their refractive index and extinction coefficient result very different from previously reported in literature. Structural, electrical and optical properties of ultrathin chromium layers are discussed in detail. The obtained optical constants of ultrathin chromium films show a specific trend with the film thickness increase. Precise knowledge of optical constants of ultrathin chromium films is important for many electro-optical and optical applications.

Keywords: chromium, sputtering, optical properties, structural properties

1. INTRODUCTION

The metal film technology is industrially important and has been improving for many years. In particular, the sputter technology developed in 18-19 century, thanks to vacuum pumps and electrical power introduction, has become one of the leading technologies for both metal and dielectric optical coatings. One of widely used types of sputtering technologies for metal deposition is magnetron sputtering. It is well known for high deposition rate at excellent repeatability and high film density.

Ultrathin metal films are widely used for optical applications, and in particular when deposited on dielectrics like glass or fused silica. The initial growth stages of ultrathin films are well known for non-linear changes in optical and structural properties of the films. The process of metal film growth on dielectrics has three stages: the formation of initial seeds and islands, the coalescence and the continuous stage. It is well known that at the initial stage, small isolated islands of metal start to form three dimensional islands on the dielectric substrate. The balance between the substrate surface energy and the energy of sputtered species determines the wetting angle of the formed islands. Hence, the type of the film growth, either Volmer-Weber or Stranski–Krastanov mode. At the first stage, metal islands are not only transparent to visible and infrared radiation, but also their density remains constant. After percolation, three dimensional islands coalesce either keeping grain boundaries between them or fusing to form a boundary free island, depending on saturation and surface energies. The final continuous stage is described by the full merge of the islands and relatively insignificant change in the film reflectance and transmittance, as well in electrical and optical properties.

The aforementioned mechanisms of the film formation have been extensively studied for noble metals like Ag, Cu and Au, while for transition metals the available literature is extremely limited. Chromium is known for fast growth of ultrathin continuous films. Precise knowledge of the film properties and optical constants of ultrathin Cr may result crucial for many electro-optical and optical applications. In this paper, the structural, electrical and optical properties of ultrathin sputtered chromium films were investigated and compared with literature data.

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2. EXPERIMENTAL DETAILS

2.1 Films preparations

For samples preparation a Kurt J. Lesker Company (PVD225) magnetron sputtering system was used. The chamber is equipped with the load lock system. As substrates, it was used double-sided polished fused silica (FS). The diameters of substrates were 25.4 mm and thicknesses were 1 mm. As a sputtering source, an unbalanced magnetron (TorusTM sputter guns) was used. It was connected to a pulsed DC (p-DC) power supply (Advanced Energy Pinnacle Plus). Power supply power was 300W. It was sputtered chromium target, 99.95% purity, manufactured by Matsurf Technologies Inc. An argon gas used for depositions was >99.999 % pure. The Ar flow rate was fixed to 20 sccm. The pumping speed of a Cryo-Torr 8 F pump was adjusted to achieve the Ar pressure of 2.2 mTorr.

2.2 Films characterization

In-situ optical transmittance was measured by broadband monitoring from Cutting Edge Coatings GmbH (CEC). The spectra were acquired in the range from 400 nm to 1000 nm through the growing film. The films were analysed for the optical transmittance and reflectance using Photon RT (Essent Optics) UV-VIS-NIR spectrophotometer and FT-IR Thermo Electron Nicolet 8700 spectrometer. To determine the optical constants of the films, ellipsometric measurements were performed using a J.A. Woolam VW-VASE instrument in the spectral range 260-2500 nm, at three incidence angles 55°, 65° and 75°. Pure metallic state of the films was confirmed together with their polycrystalline structure and low surface roughness. The density and thickness of the films were measured by XRR using a SmartLab diffractometer with Cu $K_{\alpha1}$ radiation separated by Ge(400)x2 monochromator. Furthermore, the film structure and morphology were investigated by AFM, XRD, and XPS. These last data will be illustrated in a dedicated article, while here we will only very partially notice the main results of this structural and microstructural investigation.

3. RESULTS AND DISCUSSION

3.1 In-situ change of transmittance

Firstly, we performed real-time in-situ investigation of the film growth at various wavelengths (Fig.1).

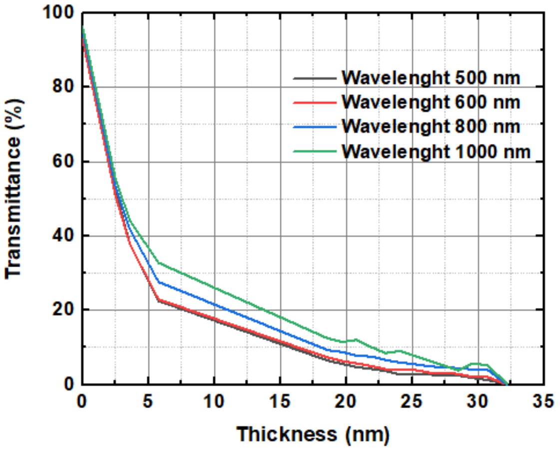

Fig.1 Real-time in-situ transmittance values changes with chromium film growth measured at various wavelengths.

It is seen that the optical properties of the growing chromium layers are not changing monotonously, yet without rapid changes, in contrast with what was previously observed for copper films [1]. For sputtered noble metals like Cu, Ag and Au, initial growing stages are characterized by a well-pronounced plasmonic peak due to the absorption in metal nanoclusters where localized surface plasmons resonances (LSPR) are excited. For such materials the percolation stage lasts longer than for chromium. Although Cr is known to be suitable for plasmonic applications, our films do not exhibit any LSPR-related feature, neither in in-situ nor in ex-situ spectra.

3.2 Ex-situ properties of sputtered ultrathinchromium thin film

The chromium film of 57 nm thickness has a density of 7.07 g/cm^3. This is close to the previously reported values of 7.11 g/cm3 and 7.2 g/cm3 [2,3]. The 57 nm sample is continuous and has and a very low transmittance. This thickest film of the sample set was used as a starting point for the characterization of the optical properties of the entire set of the ultrathin chromium films. The thickness value of 56.85 nm obtained by XRR was used as a fixed parameter enabling unequivocal determination of the film optical constants from ellipsometric spectra (Fig.2). In order to fit the acquired ellipsometric spectra, the dispersion of the refractive index and the extinction coefficient has been modeled using a superposition of Lorentz and Gaussian oscillators. A slight variation of only three parameters describing these oscillators resulted sufficient to model the optical functions of all films down to 3 nm film thickness. Details on optical and electrical characterization of the entire film set will be reported in a dedicated article.

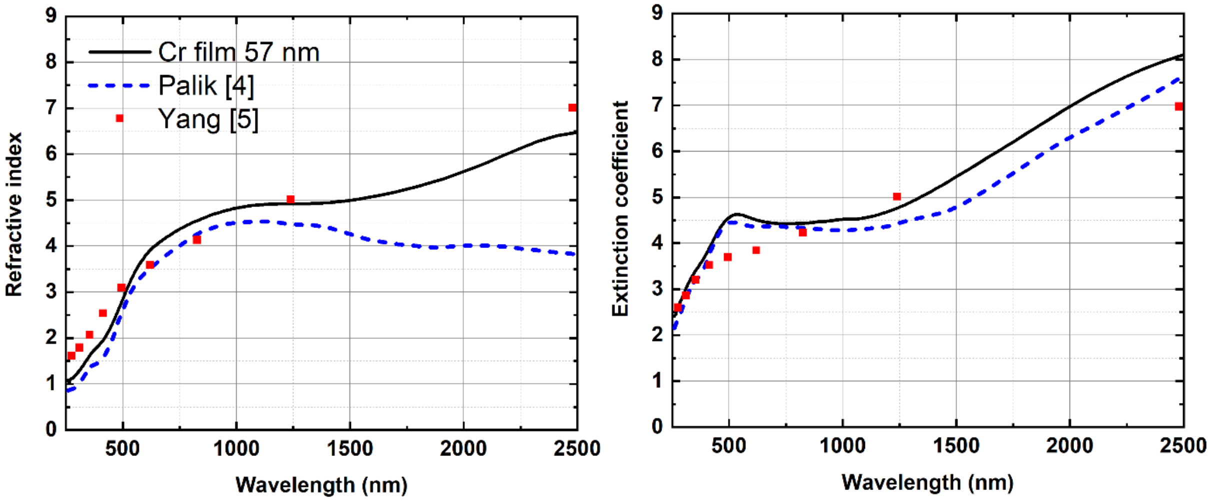

Fig.2 Evaluated 57 nm chromium film optical constants from ellipsometric measurements compared to bulky fused Cr (Palik [4]) and polycrystalline bulky Cr (Young [5]).

Low surface roughness values are important to minimize optical losses due to scattering. The root mean square roughness, R_q, was measured by AFM for 57 nm thick chromium film (Fig. 3). In this case, R_q value was about 4 Å, which is similar to the measured roughness of the bare substrate. This confirms that the roughness overlayer may be neglected for the ellipsometric data modeling.

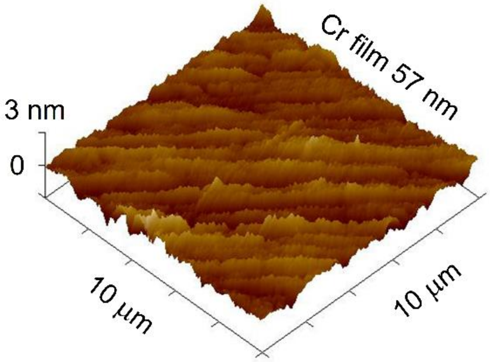

Fig.3 AFM images of 10 x 10 µm scan area of 57 nm Cr film.

4. CONCLUSIONS

Ultrathin chromium films were deposited by magnetron sputtering. The results of in-situ transmittance changes are reviewed. The density of the thick film of approximately 57 nm was 7.07 g/cm^3. In the measurement wavelength range, no evidence of LSPR excitation in the formed metal agglomerated was found. The calculated optical constants of the films are significantly higher compared to those of traditionally referenced fused bulky chromium, in particular in the infrared part of the spectrum, confirming the presence of residual boundaries between the crystallites.

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