

Sputtered molybdenum films: Structure and property evolution with film thickness



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ABSTRACT

Molybdenum thin films are widely used as e.g. diffusion barriers in very large-scale integrated circuits and thin film transistor displays. For better understanding of the evolution of structure and properties, films with different thicknesses have been grown on glass substrates by magnetron sputter deposition. A comprehensive relationship between film thickness, microstructure, stresses and resistivity could be established. With increasing film thickness, growth of (110) orientated domains is promoted, while resistivity as well as residual stress reach constant levels for films thicker than 0.7 μm .

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1. Introduction

In the last decades, multifunctional molybdenum films have gained importance in many different technological areas. They act as protective films, are applied in high-temperature applications and as diffusion barriers in microelectronics. Furthermore, they are used as an interconnect material in very large-scale integrated circuits and most commonly as back-contact layer for solar cells and thin film transistor displays [1–3]. Molybdenum is preferred because of its remarkable properties, i.e. low coefficient of thermal expansion (and therefore good adhesion to glass), good chemical and thermal stability as well as low electrical resistivity. For the applications mentioned, high throughputs demanding large-area film deposition are required. Homogeneity over large areas, reproducibility and stable, efficient deposition processes are challenges in growing these films [4,5]. Because of its capability for up-scaling to large-area deposition, magnetron sputtering is the method of choice for the synthesis of molybdenum films.

Several investigations published in literature focus on the change of properties of molybdenum films, such as morphology, internal mechanical stress and electrical resistivity, as a function of parameters of the deposition process like gas pressure or sputtering

power [4,6,7]. The aim of this study is to investigate the evolution of structure and properties of sputtered molybdenum films with film thickness, with the goal to contribute to the mechanisms of film growth and to determine the film thickness necessary for constant levels of resistivity and stress.

2. Experimental methods

Molybdenum thin films were deposited in a laboratory-scale unbalanced d.c. magnetron sputter system described in detail in an earlier publication [8]. The molybdenum target with a purity better than 99.95 wt.-% and a diameter of 152 mm was mounted to an unbalanced Gencoa PP150 magnetron cathode. The distance between target and substrate was 7.5 cm. Prior to deposition, the target was pre-sputtered for 10 min and the substrates were ion-etched at -1250 V for another 10 min to remove contaminations. While the target was water-cooled, the substrate was heated up to 200 °C during deposition. Film deposition parameters were a power density of 5.5 W cm^{-2} applied to the target, an Ar flow of 15 sccm, which led to a chamber pressure of 0.5 Pa, and a bias voltage of -50 V. The substrates, thin glass sheets (Corning EAGLE2000TM AMLCD, as used in thin film transistor display technology) of $50.8 \times 50.8 \times 0.7$ mm, have been cleaned before deposition with a commercial detergent from Borer Chemie AG. Molybdenum films with thicknesses of 0.3 , 0.5 , 1 , 3 and 5 μm were

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obtained by deposition times of 120, 180, 360, 1000 and 1800 s, respectively. A scanning electron microscope (Zeiss EVO-50) was used to determine the film thickness by investigating the fracture cross-sections. X-ray diffraction (XRD) measurements were carried out with a Siemens D500 diffractometer using Cu-K α radiation. To illuminate film growth, measurements with glancing-angle geometry (angle of incidence: 2°) were carried out. The data retrieved from Bragg-Brentano measurements were used for analysing the crystal structure as well as for calculating the coherently diffracting domain sizes and microstrains with the single-line method using a Pseudo-Voigt function [9]. The lattice distortion was calculated from the deviation of the measured with respect to the standard peak position of stress-free molybdenum [10]. Pole figure measurements were recorded using a Rigaku SmartLab 5-axis X-ray diffractometer equipped with Cu-K α radiation, parallel beam optics and secondary graphite monochromator. Residual stresses in the films were evaluated using the $\sin^2\psi$ method [11]. X-ray elastic strains were determined by measuring positions of Mo (110) reflections at different sample tilt angles ψ using a Seifert PTS 3000 four circle diffractometer [11]. The angle ψ is the angle between the sample normal and the diffraction vector. The measured lattice spacing d_{ψ}^{110} of the Mo (110) reflection and the sample tilt angle ψ were used to calculate the in-plane isotropic biaxial residual stress σ according to

$$d_{\psi}^{110} = d_0^{110} \left(1 + \sigma \left[2s_1^{110} + \frac{1}{2}s_2^{110} \sin^2 \psi \right] \right) \quad (1)$$

where s_1^{110} and $1/2s_2^{110}$ are the X-ray elastic constants (XECs) [11]. It was supposed that the residual stresses in the films are in-plane isotropic, i.e. $\sigma = \sigma_{11} = \sigma_{22}$ [12–14]. XECs for the Mo film were calculated from the single-crystal elastic constants (SECs) assuming the Hill model [15]. The electrical sheet resistivity of the films was determined with a Jandel RM2 four-point probe.

3. Results and discussion

All deposited molybdenum films adhere well to the glass substrates and show a mirror-like appearance with a bright shiny metallic surface. The cross-section of a film with a thickness of about 3 μm shown in Fig. 1 exhibits a dense and columnar structure, which is in good agreement to those films reported by Drüsedau et al. [16]. A more careful inspection reveals small crystallites formed in the early film growth stage (i.e. close to the glass substrate), whereas the later formed V-shaped columns (indicated in Fig. 1) are a result of competitive growth [17].

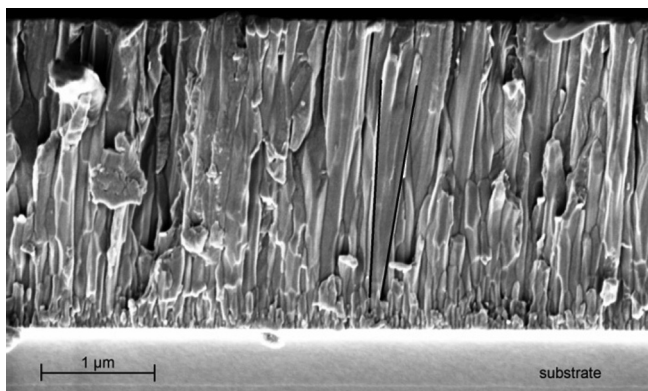


Fig. 1. SEM cross-section micrograph of a molybdenum film with a thickness of $\sim 3 \mu\text{m}$. A V-shaped column is indicated, ranging from near substrate surface to the top of the film.

The XRD patterns (Bragg-Brentano geometry) of films with different thicknesses are shown in Fig. 2a. Since the films were grown onto glass substrates having an amorphous structure, the appearing peaks only reflect the crystalline phases of the film. The peaks at $2\theta = 40.36^\circ$ and 87.6° originate from the {110} family planes of body-centred cubic molybdenum [10]. The relative intensities of the peaks originating from (110) planes increase with film thickness in contrast to the relative constant intensities collected from (211) planes (at $2\theta = 73.68^\circ$), while the peaks originating from (100) oriented lattice planes do not appear. The intensity ratio of (110) to (211) peaks and the film morphologies (Fig. 1) indicate a preferred (110) orientation, which has also been reported by Klabunde et al. [6] for low pressure film growth. The glancing-angle XRD patterns in Fig. 2b again indicate (110) oriented domains, with decreasing intensity for increasing film thickness. This is attributed to the structure evolution, with many small randomly oriented crystalline domains formed in the early growth stage and coarse columns with relatively sharp (110) orientation developing in the later stage (see Figs. 1 and 2). Reflections originating from (110) planes of these coarse V-shaped columns are rarely seen in glancing-angle geometry (see Fig. 2b for huge film thicknesses), since the diffraction vectors are not any more perpendicular to the substrate surface. In addition to these columns, (211) randomly orientated domains are formed for all film thicknesses (see Fig. 2).

To confirm the columnar structure, pole figures of the $\sim 3 \mu\text{m}$ thick film are shown in Fig. 3. The pole figure of the (110) reflection

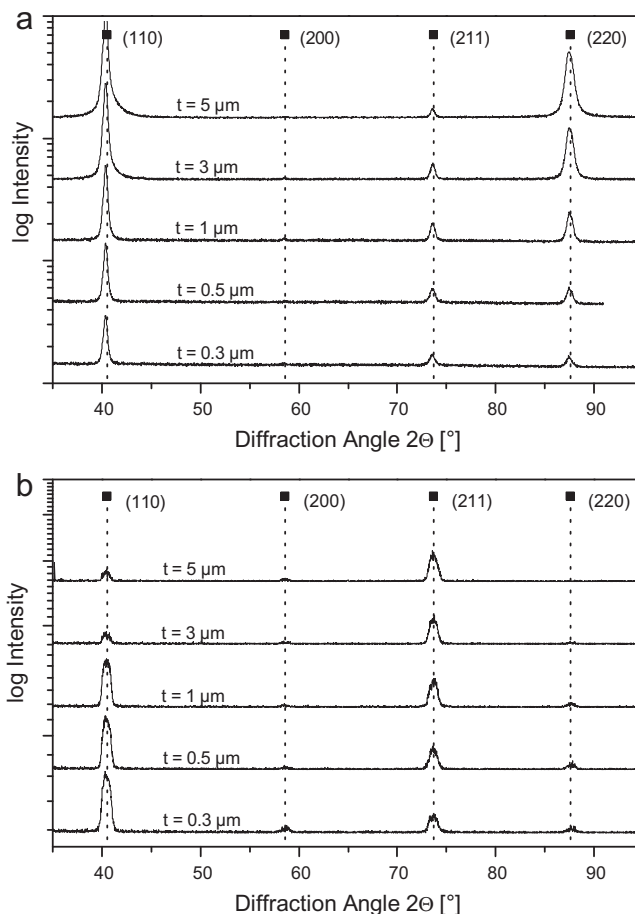


Fig. 2. XRD patterns of molybdenum films with different thicknesses obtained in (a) Bragg-Brentano and (b) glancing angle geometry (angle of incidence, 2°).

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