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Controllable residual stresses in sputtered nanostructured alpha-tantalum

A.A. Navid and A.M. Hodge*

University of Southern California, Aerospace and Mechanical Engineering Department, Los Angeles, CA 90089, USA

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Nanostructured α-Ta films were sputtered at room temperature in order to synthesize material that had either compressive or tensile stress states (−1500 to 1000 GPa). The films were coated by magnetron sputtering at various pressures using Si substrates with and without an α-Ta underlayer. The roles of the substrate/film interface, underlayer, grain size and film texture were also investigated as a function of sputtering conditions and residual stress.

Keywords: Alpha-tantalum; Nanostructure; Sputtering; Thin films

Tantalum thin films have attracted considerable interest due to their diverse properties, which render these materials suitable for applications in the semiconductor industry and for microelectronic devices, especially as materials for integrated circuits [1–3]. Even though magnetron sputtering is one of the most common processing techniques for Ta it still presents two major challenges: high residual stresses and the presence of two different phases: (i) a stable body-centered cubic (bcc) phase, known as α-Ta and (ii) a metastable tetragonal phase, referred to as β-Ta [1–12]. The structure, electrical and mechanical properties of these two phases are vastly different and the stable α phase is preferred for many applications.

There are several strategies to control a specific phase formation in Ta such as changing the sputtering conditions [1–4,6–10], heating the substrate during deposition [6,13–15], applying a bias [1,16–18] and changing the substrate material [7,13,19–22]. It is typically reported that the metastable β-Ta is the most commonly formed phase during magnetron sputtering [8,11,15,18,21,23,24] and that it can be transformed to stable α-Ta by heating above 750 °C [6,25]. However, the addition of a heating step during deposition could lead to significant amounts of grain growth, which is likely to affect the overall stress state. For example, Clevenger et al. [6] showed that heating the sample causes a drastic change towards tensile stresses which is accompanied by a phase transformation from β to α, though the contribution of the stress state to the phase formation was not studied.

In general, it is not clear whether any particular Ta phase is favored at a given stress since current measurements have been performed either with samples that were heated [6,13–15] or biased during sputtering [1,16–18]. In this article, we present a study that goes beyond current research and focus on the correlation between a α phase formation and stress state as a function of sputtering conditions (without heating or bias); specifically with emphasis on controlling the residual stress in single-phase α-Ta at different sputtering pressures.

Ta thin films were deposited by DC magnetron sputtering using sputtering pressures ranging from 0.3 to 1.4 Pa at a power of 100 W on 250 μm thick Si(1 0 0) substrates. The base pressure was maintained at less than 2×10^{-5} Pa using argon (99.999%) as the sputtering gas. Films were processed with different thicknesses (the maximum thickness being 500 nm) using a 99.995% Ta target. Residual stresses were determined from wafer curvature measurements using an XP-2 stylus profilometer (Ambios, Santa Cruz, CA). The curvatures of the samples before and after deposition were measured and the specimen curvature (ΔK) was related to the film stress according to the modified Stoney's formula [26] as follows:

$$\Delta K = \frac{6\sigma h_f}{M_s h_s^2} \tag{1}$$

* Corresponding author. Tel.: +1 213 740 4225; fax: +1 213 740 8071; e-mail: ahodge@usc.edu

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where σ is the average film stress, h_f and h_s are the film and substrate thicknesses, respectively, and M_s is the biaxial modulus of the substrate. The phase formation of as-deposited Ta film was characterized by X-ray diffraction in an Ultima-IV diffractometer (Rigaku, Tokyo, Japan) using Cu Kα radiation with a wavelength of 1.5418 Å.

In Figure 1a the phase formation is presented as a function of sputtering pressure for selected pressures ranging from 0.3 to 1.4 Pa. The X-ray spectra of the films demonstrate the formation of (1 1 0) α-Ta at 0.7 Pa sputtering pressure. Under any other sputtering condition (in the 0.3–1.4 Pa range) either randomly oriented β-Ta or a mixture of α and β phases are indentified. Figure 1a shows that the film texture can change significantly by increasing the pressure. At low sputtering pressure, the (4 0 0) β-Ta peak is observed, while at 1.4 Pa sputtering pressure, a mix of α and β phases with a (2 0 2) texture is developed. These changes in β texture could be due to the various structures of β-Ta as presented by Jiang et al. [9]. In order to investigate the α-Ta formation, transmission electron microscopy (TEM) images of a typical α-Ta microstructure sputtered at 0.7 Pa were obtained using a Tecnai F30 microscope (FEI, Tokyo, Japan) and are presented in Figure 1b–d. Figure 1b shows a TEM cross-sectional image; the sample was prepared by focus ion beam (FIB) lift-out technique. It is observed that a very dense thin layer of Ta (<100 nm thick) with ~2–3 nm grain size forms starting at the film/substrate interface, and a columnar grain structure with an average column width of ~50 nm is deposited thereafter. A top view of the films is presented in Figure 1c, and confirms an average grain size of ~50 nm. The microstructure of the thin interface layer was studied by depositing a 70 nm thick Ta film on a 10 nm thick amorphous Si_3N_4 TEM substrate grid. The TEM plan view image of the interface layer is shown in Figure 1d and confirms the formation of very small grain sized Ta film on the Si substrate.

The corresponding residual stress values as a function of sputtering pressure are shown in Figure 2. It is observed that the stress increased almost linearly while increasing the sputtering pressure, and no peak in the stress trend is observed at 0.7 Pa sputtering pressure. A high compressive stress (~−1500 MPa) develops at low sputtering pressures, and as the pressure is increased to 1.4 Pa, the film grows in a high tensile stress mode (~1000 MPa). The transition from compressive to tensile stress occurs at around 1 Pa sputtering pressure. This transition in stress state while increasing the pressure has been previously reported for Ta [6,27]; however, it typically occurred at higher sputtering pressures (ranging from 3 to14 Pa) as compared to the present results. The lower transition pressure from compressive to tensile

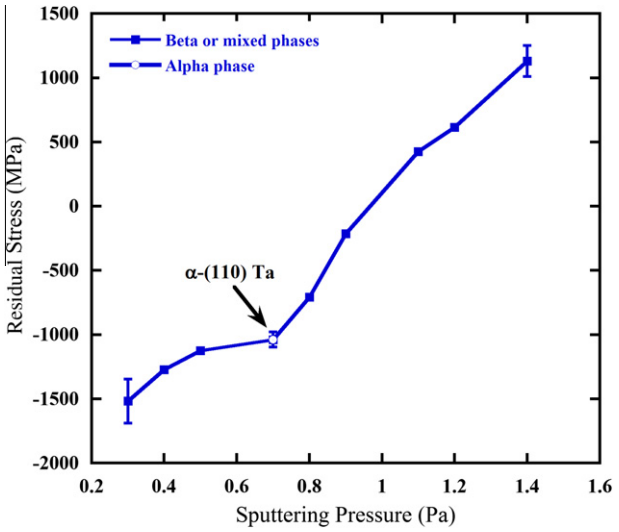

Figure 2. Plot of the residual stress as a function of sputtering pressures from 0.3 to 1.4 Pa. Arrow denotes α phase at 0.7 Pa. All other pressures have a pure β or a mixture of α and β structures.

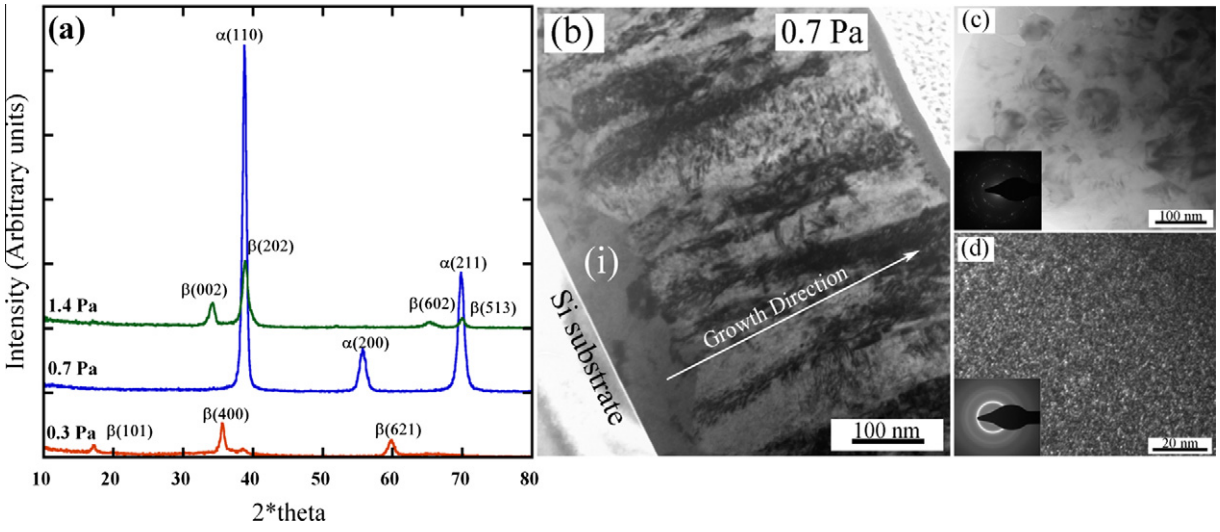

Figure 1. (a) Selected X-ray spectra of Ta films sputtered at a power of 100 W, with increasing sputtering pressures from 0.3 to 1.4 Pa. Notice the formation of α-Ta at 0.7 Pa as shown by XRD and TEM in (b)–(d). (b) Cross-sectional image, arrow denotes growth direction. (c) Top-view image. (d) Plane-view image of the interface (i) layer showing the formation of ultra-small grains at the interface between film and substrate.

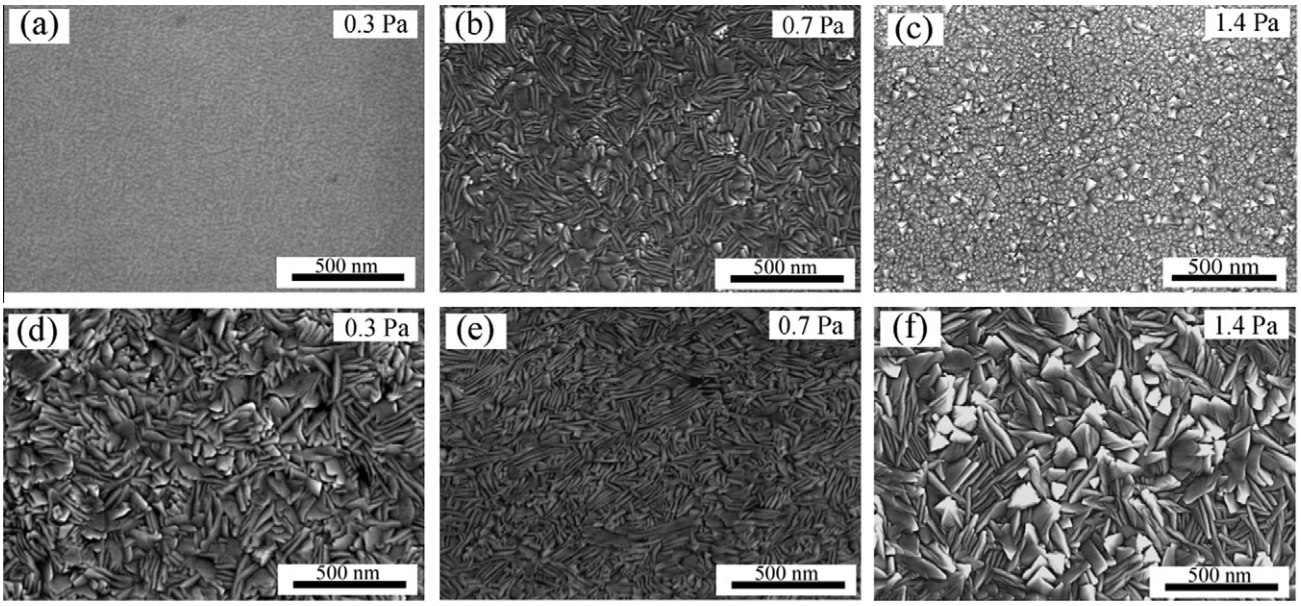

Figure 3. SEM images of the surface morphology of Ta samples synthesized at 100 W using three different sputtering pressures, 0.3, 0.7 and 1.4 Pa, deposited on two types of substrates surfaces: (a)–(c) Si substrate surface; (d)–(f) an α-Ta underlayer surface.

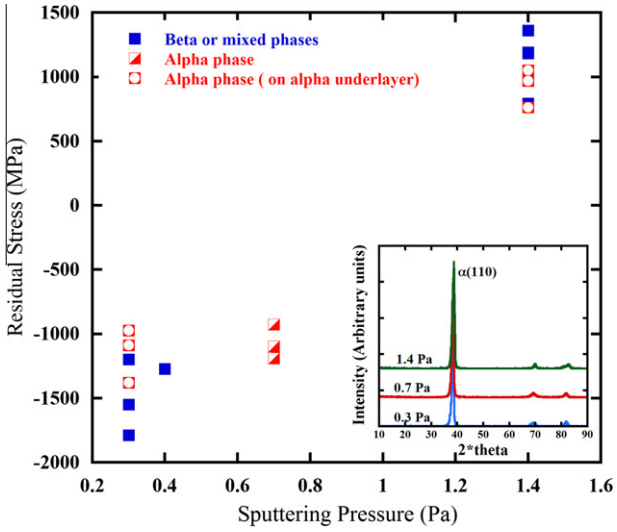

Figure 4. Plot of the residual stress of α-Ta deposited at different sputtering pressures (0.3, 0.7 and 1.4 Pa) using an α-Ta underlayer. Data points from Figure 2 are shown for reference (full square symbols). Note the formation of α-Ta at all three different sputtering pressures. Inset presents the XRD spectra of the films processed at 0.3, 0.7 and 1.4 Pa, respectively, using an α-Ta underlayer.

stress may arise from the formation of the ultra-small grain size at the interface layer (see Fig. 1d) which could promote higher tensile stress values at lower pressures [28]. Our initial results demonstrate that α-Ta developed only in a compressive mode, while β-Ta could be formed in both compressive and tensile states as the pressure is increased. At this stage it is still not obvious whether a compressive state is a prerequisite for α phase formation.

To further investigate the relationship between phase formation and stress development, we selected sputtering conditions that had yielded a high compressive or a high tensile stress (see Fig. 2) and sputtered new films using (1 1 0) α-Ta as an underlayer (see Fig. 1b). It is well established that certain substrates promote the formation of α-Ta during magnetron sputtering; however, due to the complexity of the sputtering process the effect of an underlayer is not consistent and has not been studied as a function of stress [7,13,19–21]. Therefore, Ta films were deposited at 0.7 Pa sputtering pressure to a thickness of 500 nm. Immediately after the first 500 nm were deposited, the source shutter was closed for 5 min, and then sputtering continued at either 0.3 or 1.4 Pa sputtering pressures until an additional 500 nm were deposited. The thickness of the underlayer was kept at 500 nm to avoid any probable effect of the interface layer as shown in Figure 1d. It should be noted that both 0.3 and 1.4 Pa sputtering pressures initially led to β-Ta formation as shown in Figure 2.

The surface morphologies of the films with and without α-Ta underlayer (Fig. 3) were characterized using high-resolution scanning electron microscopy (SEM) (Nova 600 Dual beam FIB, FEI, Tokyo, Japan). A typical surface morphology for α-Ta is characterized by needle-shaped particles, while β-Ta is described as having spherically shaped particles [8,18,24]. It is noted that for the samples without an α-Ta underlayer (Fig. 3a–c), both β-Ta and mixed phases were present at 0.3 and 1.4 Pa sputtering pressures, while at 0.7 Pa only needle-shaped α-Ta is present. The surface morphologies of the samples with an α-Ta underlayer are shown in Figure 3d–f. For the samples processed at 0.3 and 0.7 Pa sputtering pressure, the surface morphology has a smooth appearance which is typical for compressive films (see Fig. 2); while increasing the pressure to 1.4 Pa, results in a higher surface roughness, which is characteristic of a film under tensile stress [30].

To further elucidate the effect of the α underlayer on the film texture, phase formation and overall stress is presented in Figure 4. It should be noted that the stress values for the samples sputtered on an α underlayer were

calculated by subtracting the curvature values from the α-Ta on Si (presented in Fig. 2) from the final curvature profile of the 1000 nm thick film. Overall, Figure 4 shows that α-Ta can be synthesized in both a compressive mode at low sputtering pressures (0.3 Pa) as well as in a tensile mode at high sputtering pressures (1.4 Pa), thus indicating that no particular phase is favored for a given stress state. These results are mainly attributed to the use of an underlayer with a (1 1 0) texture which serves as a seed for further (1 1 0) growth as validated by XRD results at three different sputtering pressures: 0.3, 0.7 and 1.4 Pa (see Fig. 4 inset). Previous studies have demonstrated a similar growth effect due to a (1 1 0) texture alignment with the underlayer material given that (1 1 0) is the lowest energy lattice site configuration for a bcc material [13,29]. However, no previous studies have demonstrated the relationship between texture, phase and stress state. In general, this study has shown that having the α underlayer is a key parameter for synthesizing α-Ta with a wide range of stress values. However, which sputtering conditions always lead to the formation of the initial α-Ta underlayer is still under debate in the research community and will be the focus of a future study.

In summary, this study has shown that having an α-Ta underlayer plays a dominant role in developing an α structure (without heat or bias), since it has the same lattice parameter and the lowest surface energy texture alignment (1 1 0). It is observed that the stress evolution in Ta films can be independent of formation of a particular phase. Therefore, by manipulating the film growth by the selection of an adequate underlayer, nanostructured α-Ta can be synthesized in both compressive and tensile states, thus allowing maximum flexibility for future Ta thin film applications.

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