Determination of film stresses during sputter deposition using an *in situ* probe

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Previous work has established that the internal stresses and physical properties of sputter-deposited metals can be markedly affected by relatively small changes in the sputtering process. For instance, lowering the gas pressure or raising the discharge current can reverse the stress from tensile to compressive and, at the same time, produce more bulklike properties in the resultant coatings. Consequently, film stress observations can be utilized to map out the nature of the material produced in a given coating process and locate the boundary of the bulklike deposition regime as a function of process parameters. A principal obstacle to the implementation of this technique has been the laborious procedure of generating individual samples over a matrix of different process conditions. Residual film stresses were then determined by observing the resulting curvature of the thin substrates. In the present work substrate deflection was sensed capacitively using a miniature probe that was inserted into the deposition chamber and monitored in a remote manner. The probe signal was linear with film thickness, enabling detection of intrinsic stress in molybdenum layers as thin as 30 nm with sensitivity undiminished by multiple applications. The known compressive-to-tensile stress transition was accurately reproduced by the probe with significant additional detail. Effects of discharge current, source proximity, and argon flow conditions in shifting the stress transition were readily mapped out.

I. INTRODUCTION

Residual stresses in vapor deposited materials have been studied with many techniques, as reviewed by Hoffman[1] and by Campbell.[2] The techniques can be divided into those carried out by remote measurements within the deposition chamber and those carried out after removal from the deposition environment. While *in situ* observations are experimentally more demanding, they facilitate the examination of significant behavioral details. For instance Klokholm and Berry monitored the restoring force on a cantilever-beam substrate to establish the general constancy of intrinsic (nonthermal) residual stresses as a function of thickness in evaporated fourth and fifth period transition metals deposited at temperatures below 45% of their melting points.[3] The dependence of constant-stress behavior on substrate temperature and resulting grain size was investigated by Doljack and Hoffman by optically monitoring the curvature of a thin disk substrate suspended on a unique center-post support that also stabilized its temperature.[4] Kinbara and coworkers utilized capacitive monitoring of cantilever-beam deflection to observe the extent of constant stress-versus-thickness behavior during the deposition of thermally evaporated boron, carbon, magnesium fluoride, and titanium carbide.[5] Various other *in situ* methods and applications thereof for stress measurement in evaporated materials have been reported, although the generally simpler *ex situ* techniques are more widely used.[1,2]

In situ observations of stresses in sputter-deposited coatings are comparatively rare, attesting perhaps to the difficulty of isolating the stress transducer from the energetic sputtering environment. Stuart followed the deflection of a cantilever beam to establish that compressive stresses can occur without substrate bias in sputtered pure metals deposited at sufficiently low gas pressures.[6] Shoji and Nagata employed a similar technique to correlate the presence of compressive stress in sputter-deposited metals with the incorporation of gas.[7]

Recently the marked effects of sputtering process parameters other than substrate bias on stresses in sputter-deposited materials were further explored in a series of investigations[8–11] that employed the *ex situ* substrate bending method developed by Finegan and Hoffman.[12] This *ex situ* approach was well-suited for these investigations because it allowed the experimental samples to be generated at one site and then shipped to a distant location for analysis. In this manner it was established that the stress mapping of deposition conditions for the attainment of compressive stresses also determines the regime wherein bulklike coating properties can best be obtained. While relatively foolproof and easy to practice, the *ex situ* approach has in turn the disadvantages of being cumbersome and insensitive to the finer details of behavior. Thus, mapping out the stress effects of the numerous sputter-deposition parameters in the studies under discussion required the generation, handling, and analysis of many samples. It is to survey more rapidly the territory uncovered by the *ex situ* observations that an *in situ* approach has now been adopted for further work on the stresses and properties of sputter-deposited materials.

Like the interferometic *ex situ* method, the present *in situ* method measures the distortion or bending of a thin substrate caused by the deposition of a stressed coating, the *in situ* observation, however, being capacitive. Capacitive detection for this purpose is a documented technique,[1,2,5] although the present method differs in using a miniaturized sensor that is easily positioned and manipulated in the deposition chamber, and which is relatively insensitive to temperature. The sensor package used in this work was less than 50 mm in its greatest dimension. While the specifics of construction await separate publication, this paper will examine the type of detailed information on residual stresses in sputter-deposited coatings obtainable with such a probe and the further mapping over process parameters that has thus far been accomplished.

II. EXPERIMENTAL PROCEDURE

Experimental depositions were carried out in a 72 l, cylindrical, diffusion-pumped chamber evacuated through a cryogenic trap to a background pressure 0.05 mPa. The magnetron cathode consisted of a 0.03 m o.d. $\times$ 0.43 m long thick-walled tube of molybdenum metal inserted through a seal at the top of the chamber and closed at its lower end to contain cooling water and a magnet. The magnet was a novel construction that projected a closed-loop magnetic confinement tunnel over the outer surface of the molybdenum tube in the shape of a helix. The associated sputtering discharge formed a helical loop that was scanned around the circumference of the source by rotation of the internal magnet.[13] Thus a target area 0.33 m long on the surface of the molybdenum tube was uniformly eroded by sputtering. The maximum magnetic field strength at the target surface was 35 mTorr, and the speed of magnet rotation was 240 rpm. The cathode operated in the range of 300–600 V for argon pressures of 0.2–2 Pa (1.5–15 mTorr) and currents up to 12 A emitting molybdenum vapor at rates up to 0.6 g/min in proportion to the current.[10,11] Argon pressures were measured with a capacitance manometer located at the periphery of the deposition chamber. The capacitive film stress probe was supported on a water cooled fixture near midlength along the cylindrical magnetron cathode and at two different radial distances, 0.114 and 0.241 m, in order to determine the effect of proximity to the sputtering source. A shutter was interposed to control deposition upon the probe. In preparation for experimental depositions and observations of stress, the source was operated repeatedly at high levels of power to stimulate off-gassing and reduce the rate-of-pressure rise in the sealed-off chamber to 4×10^{-9} Pa m^3/s. When the high vacuum valve was closed, observation with a residual gas analyzer showed the partial pressures of nitrogen and water vapor to be in the range of 10^{-5} Pa with other gaseous contaminants at lower levels. In this condition the chamber could be sealed off at a chosen pressure of argon and sputtering performed with no noticeable evidence of residual gas evolution.

Rates of molybdenum deposition upon the stress probe were calibrated by reference samples on glass substrates that were measured externally with a beta-backscatter instrument. Thereafter, the extent of individual depositions upon the probe at various levels of sputtering power were in most cases inversely timed using the shutter to yield film increments of approximately 30 nm constant thickness. In practice a series of such depositions would be carried out sequentially allowing a set equilibration cycle, say 10 min, between depositions. The net shift or offset of the stress probe output in volts recorded at the end of each cycle immediately prior to the next sequential deposition was taken to be proportional to the integrated residual stress (stress-thickness product) in the preceding film deposition.

Since the incident energy per atom of sputter-deposited molybdenum has previously been measured to be about 50 eV per atom and relatively independent of deposition rate in cylindrical-post magnetron sputtering,[10,11,13] it is possible to estimate the temperature rise experienced by the stress probe during a given deposition. Heat flow calculations for a radially cooled body with uniform heating[14] indicate that the substrate portion of the probe, having an area-to-thickness ratio of less than 500 nm, is in excellent thermal contact with its surroundings, deviating therefrom in temperature by < 1 °C during the most rapid of the current depositions. Consequently, the total heat capacity of the stress probe body was used to estimate an adiabatic temperature rise per deposition of 2 °C. The effects of temperature changes of this small magnitude upon the observed stresses in molybdenum are negligible and will be disregarded for the remainder of this paper.

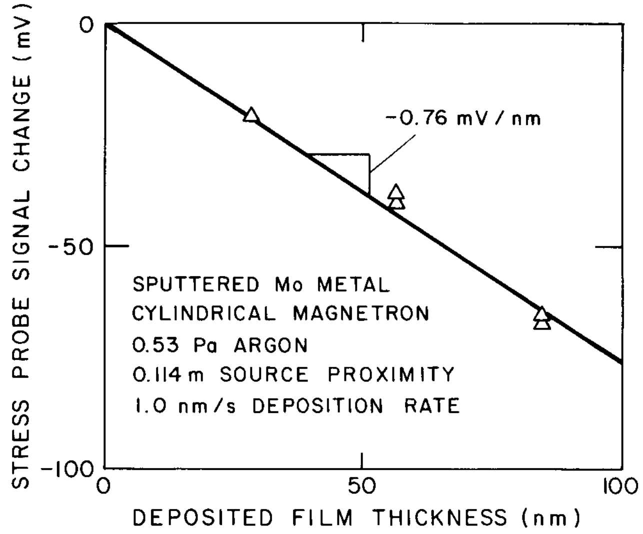

FIG. 1. Incremental output of capacitive film stress probe vs deposited layer thickness for molybdenum sputtered in 0.53 Pa (4.0 mTorr) argon at 4 A discharge current from a cylindrical magnetron source with the probe at a radial distance of 0.114 m with argon flowing at 10 std.ml/min.

III. EXPERIMENTAL RESULTS

Deviating temporarily from the procedure outlined above, films of varying thickness were deposited to determine the corresponding dependence of the stress probe response. Process parameters for these depositions were argon flow of 10 std.ml/min, pressure 0.533 Pa (4 mTorr), discharge current 4.0 A, voltage 388 V, and source proximity 0.114 m. The intent in holding these parameters constant was to fix the state of internal stress in the resulting films while varying only their thickness. The results are plotted in Fig. 1, where each point represents the net reaction of the probe to the deposition of an additional layer of thickness as indicated. The response appears to be substantially linear.

In Fig. 2 it becomes clear that the probe is responding to film stress and not, for instance, just the accumulated mass. Here as in all the data to follow the individual layers of deposition were controlled at a nominally constant thickness of 30 nm each. The probe voltage output was divided by the layer thickness to give in effect the slopes of curves like those in Fig. 1, which were then plotted. Figure 2 shows the changes in probe response accompanying an excursion in

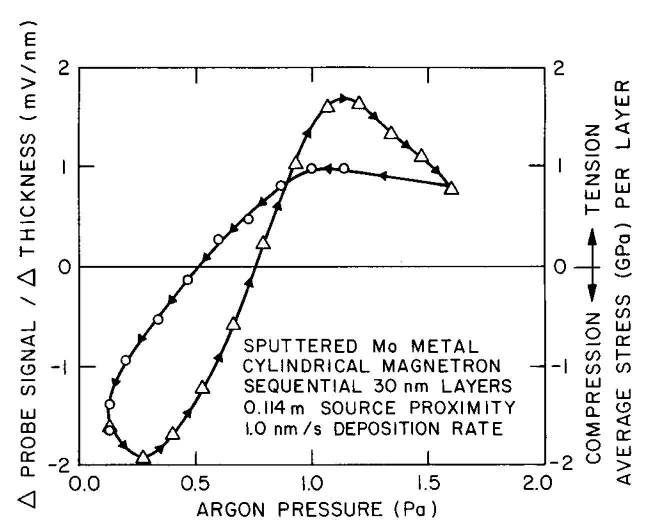

FIG. 2. Hysteresis of intrinsic film stress deposited sequentially onto probe vs argon pressure in increasing and decreasing pressure steps for molybdenum sputtered at 4 A discharge current from a cylindrical magnetron source with the probe at a radial distance of 0.114 m and with argon flowing at 10 std.ml/min.

argon sputtering pressure, first increasing and then decreasing. The result has the shape of a hysteresis loop. Consider the probe readings for increasing steps of argon pressure. These data exhibit a reversal in sign, increasing then to a maximum beyond which the magnitudes tail off gradually toward zero. The distinctive shape of this data curve corresponds closely to the well established behavior of intrinsic stresses in sputter deposited refractory metals.[8–11] It is evident from these data that the probe does indeed register the stresses in the deposited films. Further depositions in steps of decreasing pressure yielded a modified behavior lacking the distinct peak and exhibiting a more gradual transition back to negative values, which was also displaced to lower pressures. Repeated deposition at the lowest pressure produced a final negative increment that closed the experimental hysteresis loop.

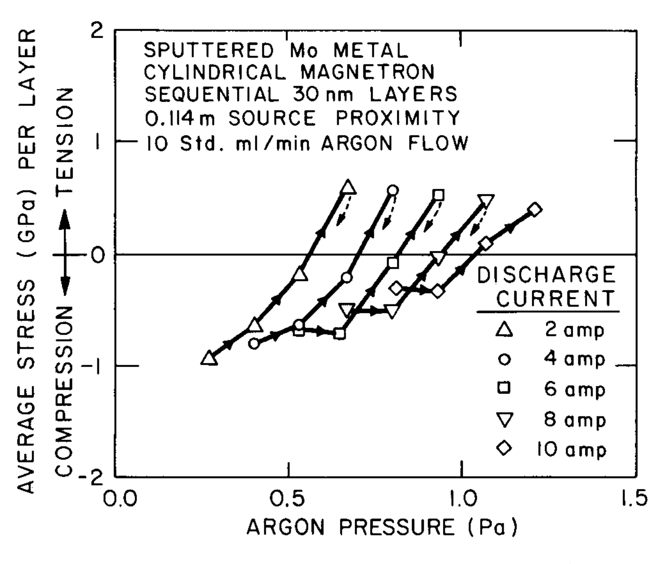

FIG. 3. Intrinsic film stress vs increasing argon pressure at discharge currents from 2–10 A for molybdenum sputtered from a cylindrical magnetron source with the stress probe at a radial distance of 0.114 m and argon flowing at 10 std.ml/min.

The correspondence between the documented tensile stress peak and the peak in the stress probe readings for steps of increasing pressure enables a rough calibration of the stress probe that is sufficient for the present purpose. In particular, the maximum in Fig. 2 is taken to be equal to the average, 1.68 ± 0.20 GPa, of three different values of the maximum tensile film stresses measured in sputtered molybdenum by the *ex situ* interferometric plate bending method[12] and reported in Refs. 10. This empirical calibration is indicated on the right margin of Fig. 2 and will serve the objective of locating the stress transition boundaries where the residual stresses pass through zero as a function of sputtering process variables, which should, in fact, be independent of the calibrating scale factor. To avoid confusion it should be mentioned that the equivalence of numerical units on the right and left sides of Fig. 2 is purely coincidental, an accident of the calibration for this particular case.

The hysteresis behavior observed in Fig. 2 implies both a persistence of established film growth structures corresponding to the varying levels of stress, yet the ability of the deposition mechanisms to overcome the currently established structure in either the compressive-to-tensile or tensile-to-compressive direction. This hysteresis also poses an experimental difficulty in working with the stress probe, as it complicates the task of reproducibly determining the stress transition pressures under various operating conditions and causes some scatter in the results that will be seen later.

Figure 3 shows results from a set of depositions aimed at determining the effect of increasing discharge current on the stress transition pressure at a selected argon flow rate (10 std.ml/min) and proximity to the sputtering source (0.114 m). The depositions were made in sequence from low to high pressure at each of the increasing levels of current until the stress crossed the zero axis from compression to tension. The previously reported shift of the transition behavior to higher pressures with increasing currents is evident.[10,11,15] Although this is a well-behaved data set, the presence of hysteresis is seen at the lower ends of the individual curves, where the tensile structure from the preceding deposition appears to moderate the magnitude of compressive stress achieved on the first deposition of the next-higher-current series. The points at which the curves of Fig. 3 cross the zero stress axis are defined to be the characteristic stress transition pressures.

Figure 4 displays the transition pressures from Fig. 3 and other similar sets of data. The scatter evident in some of these results arises from less effective control of the hysteresis than in the case of Fig. 3, which yields the uppermost curve of Fig. 4. The four conditions represented here correspond to sputter deposition with static versus dynamic argon environments at each of two different stress probe positions, i.e., proximities to the sputtering source. Despite the scatter, several immediate conclusions are obtained. The shift of transition pressure with discharge current appears to be more nearly linear than logarithmic, as proposed earlier.[10,11,15] While the shift is a general phenomenon, it is less pronounced in a static or sealed-chamber argon environment.

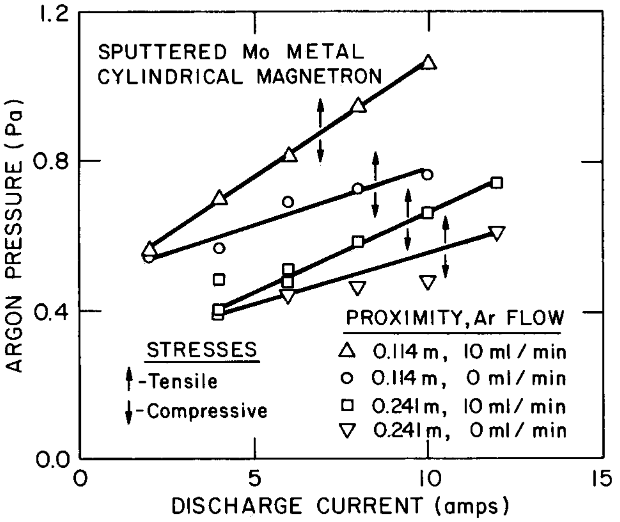

FIG. 4. Stress transition pressure vs discharge current for molybdenum sputtered from a cylindrical magnetron source with the stress probe at radial distances of 0.114 and 0.241 m and argon flowing at 10 std.ml/min and 0 std.ml/min (sealed chamber).

The stress transition pressures are displaced to lower values when the substrate is relocated farther from the sputtering source.

The compressive-to-tensile stress transition phenomenon in sputter-deposited materials has been thought to be caused by the gas scattering of particles between the source and the substrate, which increases as the gas pressure is raised. Since the lengths of intercollisional paths in a gas vary inversely with the pressure, it has been suggested that the product of pressure and source-to-substrate distance should be a generalized parameter with which to rationalize experimental observations.[16] Accordingly, the data of Fig. 4 were replotted in Fig. 5 using such a parameter. However, no evident condensation of data results from this manipulation.

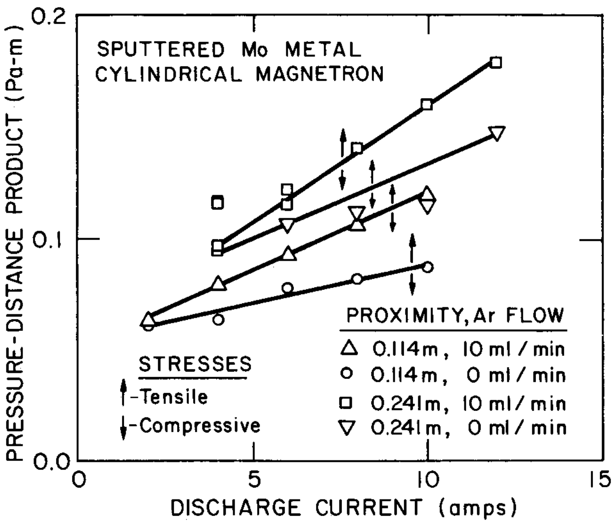

FIG. 5. Product of stress transition pressure and source-to-substrate distance vs deposition rate for molybdenum sputtered from a cylindrical magnetron source with the stress probe at radial distances of 0.114 and 0.241 m and argon flowing at 10 std.ml/min and 0 std.ml/min (sealed chamber).

IV. DISCUSSION

In the course of this work more than two hundred 30 nm layers of molybdenum were sputtered onto the film stress probe without significantly diminishing its response. A probable factor in the longevity of the probe was the careful sequencing of deposition conditions to strike an overall balance of compressive and tensile stresses in the accumulated layers, thereby maintaining an average or integrated stress of nearly zero. The long life of the stress probe in combination with its compactness suggest that it could be conveniently employed to map the types of deposition (bulklike properties or otherwise, as desired) obtainable in a given coating application as a function of process variables, and thereafter to monitor and recalibrate the process as a function of time in order to offset such gradually changing factors as sputtering target erosion, chamber contamination, etc.

The hysteresis results presented above indicate that different substrates or starting points for film deposition exert only a transient influence on the structure and properties of a given coating, which soon tends toward the characteristics dictated by the conditions of deposition alone. In particular it is significant that a film growing with tensile stress, nonbulklike properties, and, it is thought, an open structure containing atomic-scale voids can evidently be "healed" by adjusting the process parameters into the bulklike deposition regime. Finally, it is apparent that the stress transition shifts associated with increasing discharge current or deposition rate are smaller when the chamber is sealed (zero gas flow) so that the quantity of contained working gas is fixed. This smaller but significant effect, uncoupled from the changes in overall chamber pressure that occur when gas is flowing, must then relate to basic discharge mechanisms such as the observed rise in discharge voltage with rate, which may in turn reflect the dynamic rarefaction of gas near the sputtering target.[17]

V. CONCLUSIONS

(1) A miniature capacitive *in situ* probe has been employed to detect intrinsic residual stresses in sputtered molybdenum films as thin as 30 nm. The response of the probe was linear with film thickness and remained so after many accumulated layers of deposition.

(2) Intrinsic stresses observed with the *in situ* probe as a function of increasing argon pressure exhibit compressive-to-tensile transition behavior identical to that documented in the past by *ex situ* measurements. Stresses observed as a function of decreasing pressure exhibit the reverse transition but with a hysteresis that implies both "persistence" and "healing" of the film growth microstructure.

(3) The probe was used to map out the tendencies of sputtering rate, source proximity, and argon flow condition (static versus dynamic) to shift the compressive-to-tensile stress transition pressure. Regimes for deposition of films with bulklike physical properties were thereby identified.

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