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Interface creation and stress dynamics in plasma-deposited silicon dioxide films

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The stress in amorphous silicon dioxide film grown by plasma-assisted deposition was investigated both during and after film growth for continuously and intermittently deposited films. It is shown that an intermittent deposition leads to the creation of interfacial regions during film growth, but also causes dynamical structural change in already-deposited film which results in a significantly different stress-thickness profile measured after deposition. Film growth in the continuously deposited film was also monitored using an *in situ* laser reflection technique, and a strong change in stress was detected at about 145 nm which was attributed to the onset of island coalescence. *© 2006 American Institute of Physics*. [DOI: 10.1063/1.2210085]

The issue of stress in film-based structures is one of the major considerations in evaluating the performance, quality, and durability of technological applications. Stress can contribute to mechanical failure in the form of film warpage, delamination and cracking.[1,2] In devices relying on optical performance, stress can contribute to technical failure by inducing multimodes, polarization shifts, and polarization dependent loss.[3–5] On the other hand, understanding the influence of stress on materials can be utilized to control birefringence in planar devices by the use of compensation layers,[6–8] or to assist the growth of crack-free thicker film layers by depositing initial layers to counterbalance strain arising from thermal mismatch of materials.[9] A knowledge of the mechanisms responsible for the generation of stress in a material can also offer insight into the material's structure,[10,11] which may assist in the understanding of the responses and behavior of a material when it is subjected to various conditions, such as the absorption of water,[12] irradiation,[13] and annealing.[14] This is particularly relevant to amorphous materials since their structure and behavior are not yet fully understood. It is therefore of both fundamental and technological interest to understand how different growth conditions can affect the stress in a film. Monitoring the evolution of stress both during and after film growth is an effective method to achieve this. Accordingly, the evolution of stress in amorphous silicon dioxide (SiO_2) films is investigated for films deposited under two different types of growth conditions, continuous and intermittent. The evolution of stress for both film growth types was investigated during film growth using a simple *in situ* laser reflection method, then after film growth by a postdeposition etch back of the film. We present here experimental evidence of interfaces created during film deposition which are not evident in a postdeposition analysis.

Silicon dioxide films of varying total thickness were deposited for the study by helicon activated reactive evaporation[15] using small pieces of 99.99% pure Si as the evaporant material. Oxygen and argon gases were used with flow rates of 40 and 4 SCCM, respectively (SCCM denotes cubic centimeter per minute at STP). During deposition, the rf power was 800 W and the pressure was typically 1 mTorr. A constant evaporation rate was maintained throughout the deposition, being monitored by two rate crystal sensors.[16] Since the temperature during deposition is maintained below 200 °C,[17] the stress in the deposited film is intrinsic and not temperature related.[1,18] To allow consistent comparison, the deposition conditions used were identical to those used for the deposition of films intended for the fabrication of optical waveguides in the laboratory.

There are two aspects to the experiment: an *in situ* aspect, whereby the evolution of stress during film growth is monitored by a laser reflection method; and an *ex situ* aspect, whereby the stress profile of the fully deposited film was obtained only after the completion of the deposition process by the employment of a postdeposition etch back with a 1:7 buffered HF solution. The etch-back method used to obtain the postdeposition stress profile has been described and employed successfully in earlier studies.[18,19] The addition of the *in situ* aspect to the established *ex situ* postdeposition etch-back method allows the comparison of stress evolution in samples grown continuously (film grown in a single, continuous deposition) and samples grown intermittently (film grown in "layers" by halting the deposition at regular intervals with venting of the system to atmospheric pressure), for greater insight into the extent of dynamics involved in film stress formation.

The geometry of the laser reflection method utilized in the *in situ* aspect of the experiment is shown in Fig. 1. Changes in the substrate curvature due to stress were monitored by a laser reflecting from the film surface onto a screen. The substrate wafer sits in a substrate holder on an alumina ring at the top of the deposition chamber. The laser beam enters through a window at the side of the deposition chamber, reflects off the surface of the wafer, and exits through a

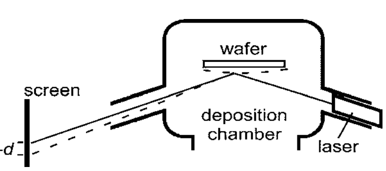

FIG. 1. Geometry of the laser reflection technique with the wafer, deposition chamber, and screen.

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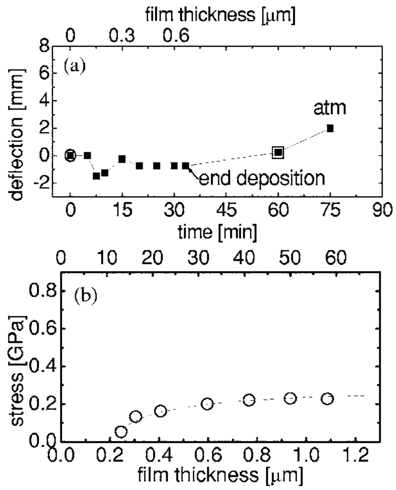

FIG. 2. *In situ* and *ex situ* results for continuous film growth: (a) Laser deflection during film growth with the initial zero reference (circle), at regular intervals during actual film growth (closed squares), system shut down and prior to venting (open square), and venting of deposition chamber to atmospheric pressure ("atm"); (b) stress profile obtained from a postdeposition etch back for a fully deposited film.

window on the opposite side of the chamber. The laser is angled at 20° to the horizontal. The horizontal distance from the center of the substrate wafer to the laser window is 140 mm and the vertical distance is 40 mm. The horizontal distance from the center of the substrate wafer to the screen is 3800 mm and the vertical distance is 1350 mm. The deflection of the laser spot on the screen, d, was marked manually at regular intervals. Thus the deflection of the laser spot as a function of time corresponds to the evolution of stress in the film as film growth is taking place. When deposition is complete, measured deflections of the laser spot provides an indication of additional, postdeposition dynamics influencing the stress in the film. The deflection of the laser spot measured just prior to the removal of the sample completes the *in situ* aspect of the experiment.

After the sample is removed, the *ex situ* aspect of the experiment proceeds with the postdeposition etch-back method,[18,19] whereby the film thickness was determined by a J. A. Woollam M-44 WVASE ellipsometer, and the bowing in the substrate was measured by a Tencor P-10 surface profiler, after film etch. The average film stress was calculated from the measured bow using Stoney's equation,[19,20] utilizing a profiler scan length of 10 mm and a substrate elastic constant of 180.5 GPa (Ref. 21) for the 4 in. ⟨100⟩ Si substrates (p type) of 0.5 mm thickness. Thus the calculated average stress as a function of film thickness corresponds to the stress profile of the film when it is etched back from its fully deposited state, representing lasting influences of any postdeposition dynamics.

Figure 2 shows the results for the continuously deposited film from the *in situ* and *ex situ* aspects of the experiment, with (a) the evolution of stress detected during film growth, represented by the vertical deflection of the laser beam as a function of time, and (b) the stress profile of the film after postdeposition etch back from its fully deposited state. In Fig. 2(a), the evolution of stress in the continuously deposited film was monitored at specific and regular intervals over the course of the entire deposition process until the sample was removed, with a downward deflection taken as negative. The maximum relative error in the measured laser spot position is ±5%. The initial position of the laser spot (open circle) represents an ~10 min period, from the time the system (plasma and e-beam) was still off until after a plasma stabilization time of 5 min. The laser spot position remained unchanged during this period which involved a pressure difference of $\sim 10^3$ Torr. During the course of film growth (closed squares), a significant negative deflection was observed at 7 min and 20 s into the film growth, which corresponds to a film thickness of about 145 nm. Coinciding with the observed negative deflection, the laser spot appeared smaller and fainter, and only returned to its original intensity and size 10 min into film growth (about 175 nm). Other than this event, a largely constant deflection was observed, with further (positive) deflections occurring only after deposition had finished, the plasma and e-beam power reduced and the entire system turned off for 10 min (open square), and after venting of the system to atmospheric pressure, just prior to removal of the sample ("atm"). The negative deflection occurring at around 145 nm film thickness is therefore considered to be the most significant event during the course of a continuous film growth process. In Fig. 2(b), this particular event was not recorded by the postdeposition etch back; however, the remaining stress behavior corresponds well to what was observed through the *in situ* treatment, i.e., a plateau in the profile towards a constant stress value. The error in the calculated stress is about ±0.1 GPa.

It is possible that this observed significant event corresponds to the onset of island coalescence in the film. In an earlier study, it was found that continuously deposited SiO_2 films grown by helicon activated reactive evaporation were described by a Volmer-Weber treatment and that a sharp increase in stress attributed to island coalescence should begin for a film thickness less than 165 nm.[18] The combination of the observed dynamics acquired during film growth in the current study and the results from the prior study on continuously deposited films by the same deposition method strengthens the argument that a significant event, such as the onset of island coalescence, occurs at a film thickness of around 145 nm.

Figure 3 shows the corresponding results for an intermittently deposited film from the *in situ* and *ex situ* aspects of the experiment. Figure 3(a) shows the evolution of stress detected during film growth and Fig. 3(b) shows the postdeposition stress profile. The intermittently deposited sample was comprised of four layers of equal thickness, with venting of the system to atmospheric pressure between each layer. Typically, deposition of subsequent layers was not resumed until the following day. It should be noted that until the venting of the system, the deposition of each layer proceeds as per a continuous deposition. The dynamics up until that point will therefore follow those observed for the continuously deposited film as shown in Fig. 2. The interest in the intermittent study, then, is the effect of introducing the intermittent process or deliberate interruption into the film growth. Hence, it is only at the end of the deposition of each layer that the position of the laser spot was marked with the corresponding layer (e.g., events "1," "2," etc.). The notation for all other events marked for the laser spot position follows that of the continuous deposition. Note that there is a period of 24 h between depositions of layers [i.e., between events atm and circles in Fig. 3(a)]. This is denoted by breaks in the horizontal (time) axis. Film deposition only occurs between the circled event and numbered events, 1, 2, etc. All other

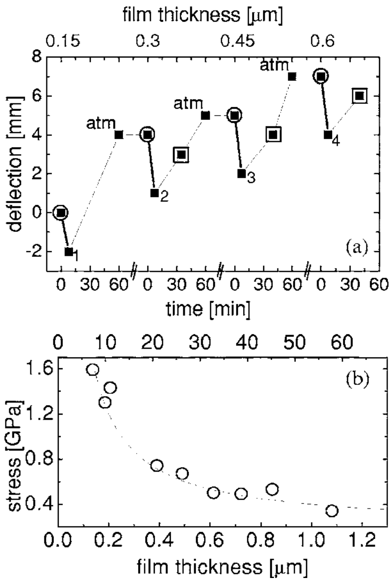

FIG. 3. *In situ* and *ex situ* results for intermittent film growth: (a) Laser deflection during film growth, deposition occurs between events first circle and "1" (for this case, 0–150 μm), second circle and "2" (150–300 μm), third circle and "3" (300–450 μm), and fourth circle to "4" (450–600 μm) (b) stress profile obtained from a postdeposition etch back for a fully deposited film.

deflections reflect dynamics occurring when the film thickness is not changing.

The strongest indication of the creation of an interface coinciding with the introduction of the intermittent process is shown in Fig. 3(a) by a change from a net *negative* displacement when a layer is completed (e.g., from circle to event 1) to a net *positive* displacement by the time the next deposition is about to commence (e.g., event 1 to next circle). It is worth noting also that the position of the laser spot remains unchanged over the 24 h period (event atm to circle) as the chamber pressure returns to deposition conditions overnight following the previous day's venting to atmospheric pressure. In contrast, the postdeposition stress profile of an intermittently deposited film [Fig. 3(b)] does not reflect the presence of any abrupt changes to indicate the presence of interfaces. The function in Fig. 3(b) is smooth, with a concentration of higher stress only existing near the film-substrate interface.

An earlier study described the structure of amorphous SiO_2 film deposited by helicon activated reactive evaporation in terms of a strained SiO_4 tetrahedral ring structure.[19] The Si–O–Si bonds in the strained structure are considered unstable because an accumulation of stress in the film can break the strained bonds. It was proposed that the additional stress introduced through intermittent depositions at interface locations could induce a domino effect of further strain in the surrounding network, which would result in an overall downwards transfer of stress into the existing film, towards the film-substrate interface. This proposed dynamic mechanism for the transfer of stress through the existing film was dependent on a postdeposition dynamic process, since the concentration of stress occurs at small film thicknesses. The interface is not introduced until later in the deposition, therefore, it is not possible for the film, during growth, to exhibit differences that only much later events would cause. Evidence for this postdeposition dynamic process appears to exist in Fig. 3(a) by way of the considerable amount of dynamics detected by the laser deflection system when there is no deposition occurring and the film thickness is not changing. Dynamic changes take place *as soon* as the system is shut down after a deposition and *before* the increase of chamber pressure (e.g., event 2 to open squared event). These dynamic changes *continue* to take place as the sample is brought to atmospheric conditions (open squared event to event atm). It appears, therefore, that significant changes related to the film's stress, and hence structure, do indeed occur after a deposition is stopped or halted. Although the same dynamics were observed in the continuously deposited film, it is of particular significance to the intermittently deposited film since further depositions are made after the occurrence of these events. Since the deposition of the next layer resumes at the same position as the last atm event, it appears that the dynamic mechanisms which take place following a halted deposition are lasting.

In conclusion, we have shown that by employing a simple *in situ* laser reflection setup, there is evidence that an intermittent deposition causes the creation of interfaces at the locations of halted film growth. The process also introduces additional stress to the amorphous film network which is then transferred through the existing film, towards the film-substrate interface, resulting in a significantly different stress-thickness profile from that obtained through a continuous film deposition. Further work should help refine the laser technique employed for this study and allow more precise information.

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[1] C. Charles and R. W. Boswell, J. Appl. Phys. **84**, 350 (1998).
[2] S. M. Hu, J. Appl. Phys. **70**, R53 (1991).
[3] Xiuli Zhao, Chunfei Li, and Y. Z. Xu, Opt. Lett. **28**, 564 (2003).
[4] M. Huang, Int. J. Solids Struct. **40**, 1615 (2003).
[5] L. Grave de Peralta, Ayrton A. Bernussi, H. Temkin, Marcus M. Borhani, and David E. Doucette, IEEE J. Quantum Electron. **39**, 874 (2003).
[6] Winnie N. Ye, Dan-Xia Xu, Siegfried Janz, Pavel Cheben, Marie-Josée Picard, Boris Lamontagne, and N. Garry Tarr, J. Lightwave Technol. **23**, 1308 (2005).
[7] D.-X. Xu, P. Cheben, D. Dalacu, A. Delâge, S. Janz, B. Lamontagne, M.-J. Picard, and W. N. Ye, Opt. Lett. **29**, 2384 (2004).
[8] A. Kilian, J. Kirchhof, B. Kuhlow, G. Przyrembel, and W. Wischmann, J. Lightwave Technol. **18**, 193 (2000).
[9] A. Krost, A. Dadgar, J. Bläsing, A. Diez, T. Hempel, S. Petzold, J. Christen, and R. Clos, Appl. Phys. Lett. **85**, 3441 (2004).
[10] L. Martinu and D. Poitras, J. Vac. Sci. Technol. A **18**, 2619 (2000).
[11] D. R. Hamann, Phys. Rev. B **55**, 14784 (1997).
[12] Norio Hirashita, Shunichi Tokitoh, and Hidetsugu Uchida, Jpn. J. Appl. Phys., Part 1 **32**, 1787 (1993).
[13] E. Snoeks, T. Weber, A. Cacciato, and A. Polman, J. Appl. Phys. **78**, 4723 (1995).
[14] R. A. B. Devine and M. Marchand, Appl. Phys. Lett. **63**, 619 (1993).
[15] A. Durandet, R. Boswell, and D. McKenzie, Rev. Sci. Instrum. **66**, 2908 (1995).
[16] W. T. Li, D. A. P. Bulla, C. Charles, R. Boswell, J. Love, and B. Luther-Davies, Thin Solid Films **419**, 82 (2002).
[17] G. Giroult-Matlakowski, C. Charles, A. Durandet, R. W. Boswell, S. Armand, H. M. Persing, A. J. Perry, P. D. Lloyd, S. R. Hyde, and D. Bogsanyi, J. Vac. Sci. Technol. A **12**, 2754 (1994).
[18] V. Au, C. Charles, D. A. P. Bulla, J. D. Love, and R. W. Boswell, J. Appl. Phys. **97**, 084912 (2005).
[19] V. Au, C. Charles, and R. W. Boswell, J. Phys. D **39**, 164 (2006).
[20] G. G. Stoney, Proc. R. Soc. London, Ser. A **82**, 172 (1909).
[21] W. A. Brantley, J. Appl. Phys. **44**, 534 (1973).