Periodic Variation of Stress in Sputter Deposited Si/WSi₂ Multilayers

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A tension increment after sputter deposition of 1 nm of WSi₂ onto sputtered Si was observed at low Ar gas pressures. Wafer curvature data on multilayers were found to have a periodic variation corresponding to the multilayer period, and this permitted statistical analyses to improve the sensitivity to small stresses. The observation of tension instead of compression in the initial stage of growth is new, and a model invoking surface rearrangement is invoked. The data also bear on an unusual surface smoothing phenomena for sputtered Si surfaces caused by the sputter deposition of WSi₂. We furthermore report that for low Ar pressures the Si layers are the predominant source of built-up stress.

Although adatom induced changes in surface facets of Si crystals has been much studied ⁴, adatom induce changes in the surfaces of amorphous Si have not been reported, nor are we aware of such studies for the surface of other amorphous material. There are a wide variety of applications for amorphous thin films, and the atomic origins of interface stress in thin films formed by sputter deposition bear on these applications ².

We report the observation of tension for the first nanometer of WSi₂ deposited on amorphous Si, and these results are counter to the often invoked lock-down mechanism which results in compressive stress. Sputtered multilayers are amorphous ³,⁴ and are ostensibly free of crystalline relaxation mechanisms such as dislocation motion and crystalline anisotropies ⁵. This makes them more ideal for the study of stress induced by local relaxation such as hybridization caused by adatoms.

As an example of a particular application, Si/WSi₂ multilayers consisting of many hundreds of periods have been used to make lenses for nanofocusing of hard x-rays.⁶,⁷. Successful focusing lenses have been made in spite of the built-up stress, but that each WSi₂ layer smoothens to a roughness less than or equal to 0.36 nm as each Si layer in a Si/WSi₂ multilayer is grown. After sputtering of 1 nm the wafer was rotated to face a laser based system for measurement of wafer curvature ¹⁰. Stoney’s equation in differential form for the change in wafer curvature, δ(1/R), for a change in film thickness, δ(t_f), is given by ¹¹.

$$\delta(1/R) = 6\sigma(1−\nu)E_t^2$$  (1)

Here R is the radius of curvature of the wafer, σ is the biaxial stress, ν is Poisson’s ratio and E is the Young’s modulus of the substrate, t_s is the substrate thickness, and t_f is the film thickness. For Si(100) the ratio (1−ν)/E is conveniently isotropic in the plane of the wafer. Curvature measurements at azimuthal angles 90 degrees apart were found to give the same results which allowed us to rule out spurious effects due to mounting stresses or due to a flat on the otherwise round wafer. Confirmation of the multilayer period thickness was obtained with standard x-ray diffractometry.

Intriguingly and counter to the conventional expectation that a heavy ion impinging on a lighter one should cause interface mixing, in previous synchrotron studies a smoothing effect was instead found ¹². The x-ray reflectivity measurements reported by Wang et al. ¹² were made in-situ and continuously as shutters to sputtering guns were open and closed. The reflectivity was found to oscillate during growth, as expected from interference effects, but the reflectivity was constant when the shutters were closed. We infer that there is no change in either the roughness, density or thickness of a grown layer with a closed shutter, since changes in any of these would affect the reflected intensity. Consequently, for the present experiments, possible changes to the sputtered layers during the time that our samples were rotated to face to the MOS system are not considered significant. The results reported by Wang et al. ¹² are that the roughness builds up to be greater than or equal to 0.36 nm rms as each Si layer in a Si/WSi₂ multilayer is grown, but that each WSi₂ layer smoothens to a roughness less...
FIG. 1. Curvature data for sputtering at 6 different Ar pressures for a total of 20 multilayer periods. Compressive changes are negative and tensile changes are positive. For the solid points, the most compression develops for 2.3 mTorr, with progressively less compression for 6, 8, 10, and 12 mTorr. The data for 18 mTorr reveal a net tension after 20 periods. (An unintended gap in the data at 2.3 mTorr is also evident. Data in this range were not used in the analyses). (Color on-line only).

than or equal to 0.27 nm rms. These results indicate that the surface morphology of a sputtered Si layer is strongly affected by the deposition of WSi$_2$ ions. The present data add the information that these changes in surface morphology result in a tension increment.

Our data reveal a systematic change from a final compression at low Ar pressures to a final tension at higher pressures. This is a well known effect attributable to island coalescence and has been reported for a wide variety of systems (see Freund and Suresh, Fig. 1.39 [2]). In the present case at Ar pressures above 6 mTorr, x-ray GISAX data has revealed sputter deposition by landing of coalesced WSi$_2$ particles, and this is consistent with an island coalescence model at higher Ar pressures [13].

The data reveal an overall gross variation with time, and a variation about an averaged curve that repeats over the multilayer period. In each case there is a region of the gross variation which is linear as shown in Fig.2 for three periods. Although previous ex-situ studies in our laboratory hinted at this variation [14], we are not aware of other reports of a systematic curvature variation over a multilayer period. A very pertinent aspect presently is that these data permitted statistical averaging to reveal weak stress effects arising from the deposition of only 1 nm of thickness. Since the data were very periodic, they could be collapsed into a time base corresponding to a single multilayer period and averaged to yield the results shown in Fig.3.

An unexpected feature of these data is the observation of tension in the first nanometer of WSi$_2$. As in the case of crystals, surface reconstructions on the surfaces of amorphous solids should be driven by an inequality between surface stress, $f$, and excess surface free energy per unit area, $\gamma$. We conclude that $(f - \gamma)$ is negative for the interface created when WSi$_2$ is sputter deposited onto sputtered Si leading to a negative biaxial film stress, i.e., a tension, given by $\sigma = (f - \gamma)/t_0$, where $t_0$ is the thickness of the surface layer.

Meade and Vanderbilt report strong tensile stresses for Si(111) surfaces covered by adatoms with tensile stresses ranging from 1.18 eV to 1.70 eV per 1 x 1 cell [16]. The effect of adatoms on a Si(111) surface was calculated by them to make a tensile stress contribution [17]. Our data permit a quantitative comparison to these results. For the observed tension increment of $6 \times 10^{-4}$ m$^{-1}$ observed at 2.3 mTorr pressure after deposition of 1 nm of WSi$_2$, we can apply Eq. 1 to yield a value of $\sigma = 1.1 \times 10^{10}$ dyne/cm$^2$. Here we applied a value of $(1 - \nu)/E = 0.554 \times 10^{-12}$ cm$^2$/dyne, for the biaxial modulus of the Si(100) substrate. The product of this stress and the thickness increment of 1 nm leads to a value 0.36 eV per 1 x 1 cell (surface area equal to 0.047 nm$^2$). This value is smaller
FIG. 2. Curvature data as a function of growth time over three periods in the linear regime. Positive changes correspond to tension and negative to compression. The thickness of the both types of layers was 5 nm grown in 5 steps of 1 nm each. The growth rate of the WSi$_2$ was slightly less than for the Si with correspondingly slightly longer sputtering time to achieve the same 5 nm thickness. The data sets at each pressure have been arbitrarily offset for clarity. Error bars represent one standard deviation for 18 curvature measurements made at each point. (Color on-line only).

than those calculated by Mead and Vanderbilt [16], and we propose that is due to the amorphous Si surface in our case.

A widely applicable model for the stress evolution of deposited layers is discussed by Freund and Suresh [2], and involves regimes of stress that change with growth thickness. A compressive stage occurs at first, followed by a tension regime. For the first stage a lock-down mechanism has been invoked [2]. The idea underlying this mechanism arises from the observation that the bond length of a small particle locked to the substrate is smaller than the same material in bulk form. This brings about compression as the layer is filled in.

Our observations of tension in the first nanometer of WSi$_2$ run counter to this model, and thus we are led to invoke the above model of surface rearrangement for underlying Si layers. We note that once the first nanometer is deposited, we do see a compressive regime as is commonly expected for a peening effect for deposition of films with higher atomic masses.

As concerns a means to minimize the build up of accumulated stress, we propose that for Si/WSi$_2$ multilayer applications that require a given period, a reduced Si layer thickness should be effective.

In summary, an unexpected tension increment for the first nanometer of WSi$_2$ deposited onto Si in sputtered multilayers is reported. This tension increment is unusual in that a compression in the first stage of growth has been reported for a variety of other systems. Because the present data were obtained with multilayers, a powerful new statistical analyses is possible that provides high sensitivity to small changes in wafer curvature resulting from incremental stress. Unusual smoothing of a sputtered Si surface by sputter deposition of WSi$_2$ was previously reported [12] indicative of Si surface rearrangement, and such a model is invoked presently. Published calculations of tension caused by adatoms for a Si(111) surface can be compared quantitatively to the present case, and although similar in magnitude, our results reveal a smaller adatom induced tension for an amorphous Si surface.

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FIG. 3. Curvature data that have been collapsed onto the time base for a single multilayer period and offset for clarity. Here the error bars correspond to plus and minus a standard deviation for averaging from 5 to 8 collapsed periods. (Color on-line only).

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