LIGO Contract No. 1063039
CSIRO Report 1
May 2004
Ion Beam Deposited Coatings

Objectives

- To establish the conditions for deposition of non-stoichiometric tantalum oxide films for use in mirrors for mechanical loss testing.
- Determination of Young's Modulus of these films.
- Determination of the effect of annealing on Young's Modulus of tantalum oxide films.

Method

Tantalum oxide films were deposited by ion beam sputtering from a tantalum metal target without using additional substrate ion bombardment. The deposition temperature was about 100ºC. Layers of tantalum oxide approx 700-800 nm thick were deposited on polished (commercial grade) homosil disks 25 mm diameter and 5 mm thick, with varying oxygen partial pressure in the deposition chamber. It is known that the oxygen partial pressure is important to obtain the correct stoichiometry of the tantalum oxide films (ie Ta_2O_5) and thus obtain the lowest as-deposited absorption loss.

The optical characteristics of the films were determined from transmission measurements in a Varian Cary 5 UV-Vis-NIR spectrophotometer. Woollam's *WVASE32* software was used to model the transmission characteristics to extract the refractive index, thickness and extinction coefficient. Modelling of the extinction coefficient is difficult and so these results must be taken as a guide only, however, simple inspection of the layers can be used to verify the presence of extremely high loss.

The stoichiometry of the films was measured both by XPS (SPECS Sage system) and by inference from modelling of the transmission spectrum using the effective medium approximation (EMA) available in the *WVASE32* software package. This latter method assumes that the non-stoichiometric film can be modelled as a film with a stoichiometric component (as Ta_2O_5) with a percentage of Ta metal representing the non-stoichiometric component. It should be noted that XPS is unlikely to give the true stoichiometry to better than a few absolute percent.

The Young's Modulus of the films was measured using a CSIRO manufactured ultra-micro indentation system (UMIS). An explanation of this system and its limitations is provided in the appendix to this report.

Results

Extinction Coefficient

The extinction coefficient of approximately 750 nm thick films of tantala as a function of the oxygen backfill flow rate is shown in Fig. 1. The backfill flow rate is proportional to the oxygen partial pressure. The absorption of the unannealed films increases rapidly with lower flow rates. While determination of high absorption values by modelling is prone to inaccuracies due to the many ways in the absorption can be modelled, the films are essentially transparent at high flow rates, but become obviously darker with decreasing flow. This can be seen in Fig. 2. In other words, the low oxygen flow rate material is very absorbing.

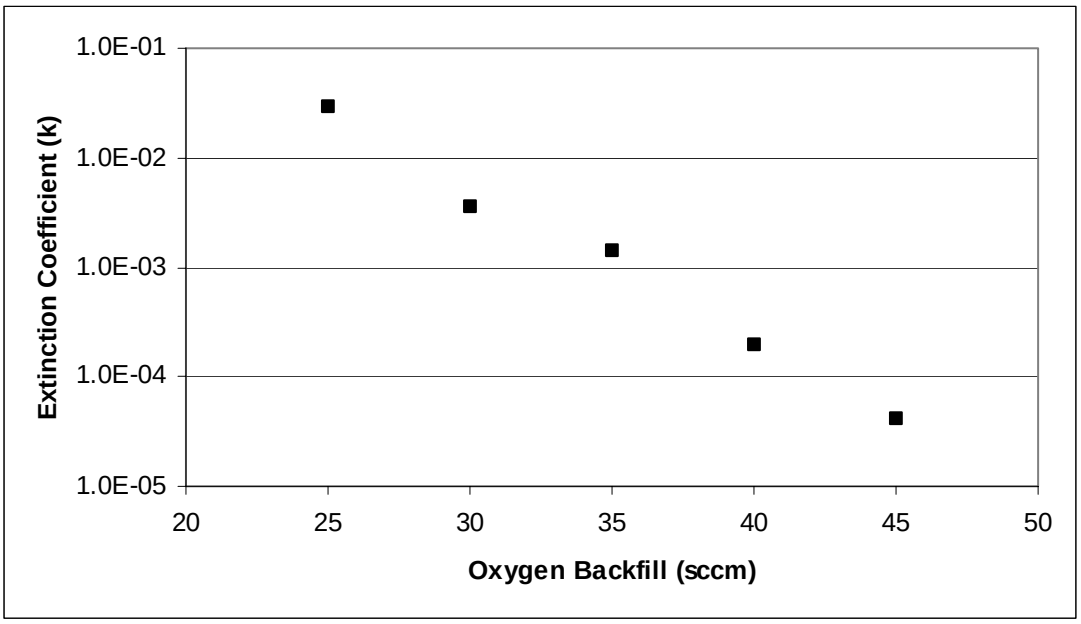

Fig. 1 Modelled extinction coefficient of unannealed tantala films (at 1064 nm) vs oxygen flow rate.

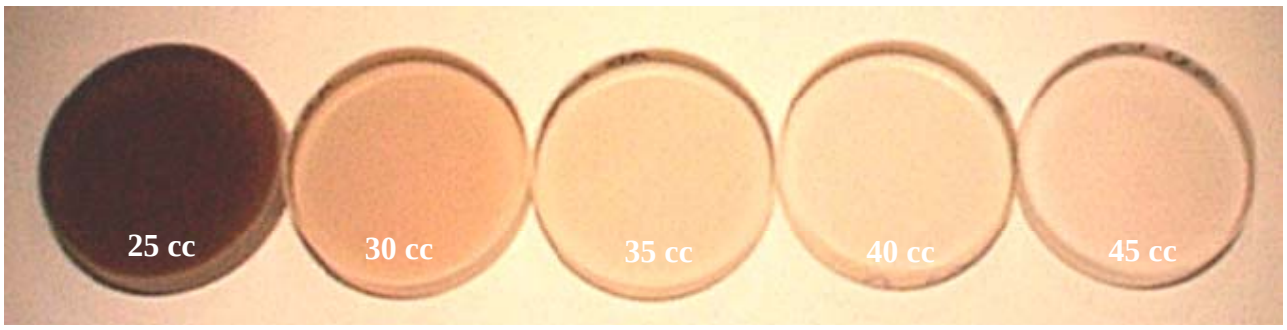

Fig 2 Physical appearance of the substrates with data in Fig. 1

Film Stoichiometry

The stoichiometry of the films, as determined by XPS, is shown in Table 1. A piece of bulk Ta_2O_5 is shown for comparison and calibration purposes. The expected value for Ta_2O_5 is Ta:O equal to 28.4:71.6 (atomic %), which can be compared to the XPS value for the bulk material of 30.1:69.5 obtained here. This sets a limit of about 5% to the accuracy of the results.

Table 1: Stoichiometry of tantala films determined by XPS measurements

	at. %	Bulk Ta_2O_5	Thin film 45 sccm O_2	Thin film 40 sccm O_2	Thin film 35 sccm O_2	Thin film 30 sccm O_2	Thin film 25 sccm O_2
As deposited	O	69.5	69.8	69.6	69.5	69.9	69.1
	Ta	30.1	30.2	30.4	30.5	30.1	30.9
Etched for 1min Ar@5kV, 2mA	O		69.8	69.8	69.2	69.2	68.7
P 5*10-3mbarr	Ta		30.2	30.2	30.8	30.8	31.3
Etched for 5min Ar@5kV, 5mA	O	66.9	69.1				62.8
P 5*10-3mbarr	Ta	33.1	30.9				37.2

All of the as-deposited films show a stoichiometry approximately the same as the standard, despite different deposition conditions and optical appearance. This is unsurprising since any unsatisfied bonds near the sample surface are likely to be fully oxidized upon exposure to air. As XPS measures film properties only down to ~4-5 nm depth, it is unlikely to 'see' beyond this native oxide. Ion beam etching of the films was thus carried out in the XPS system to remove any native oxides. Unfortunately, this process itself can result in changes in the stoichiometry of the film due to preferential sputtering and ion-induced mixing. This can be seen in the change in apparent stoichiometry of the bulk sample following ion etching. There is also a change in the stoichiometry of the sample deposited with 45 sccm of oxygen backfill, but it is much smaller than that of the bulk material, perhaps because the latter is a sintered powder and the film is a continuous dense layer. The change in Ta content in bulk tantala can therefore be taken as an upper limit of the degree of ion-etch distortion of the stoichiometry and is about 10% (3% absolute). Despite these shortcomings, it can be seen that the sample deposited with 25 sccm of oxygen backfill, has a stoichiometry of Ta:O of 37:63 which is a much higher Ta content than might be explained by preferential sputtering. We can combine these results to infer that the Ta content has increased in this case by between 10% and 20% (3%-6% absolute).

A corroborating estimate of the tantalum content can be made by modelling the transmission of the film using an effective medium approximation (EMA). Here the film is assumed to be composed of Ta_2O_5 and Ta and the transmission calculated as a

weighted average of the optical constants of the two materials. Performing this calculation in WVASE for the 25 sccm oxygen backfill sample, a similar transmission profile can be obtained for a relative increase in Ta content of he film of about 6-10%, which is similar to the XPS result.

Young's Modulus

Figure 3 shows the elastic (Young's) modulus as a function of probe penetration depth for the various oxygen backfill flow rates used previously. The sample number is shown instead of the flow rate, but it can be seen that the results are essentially indistinguishable.

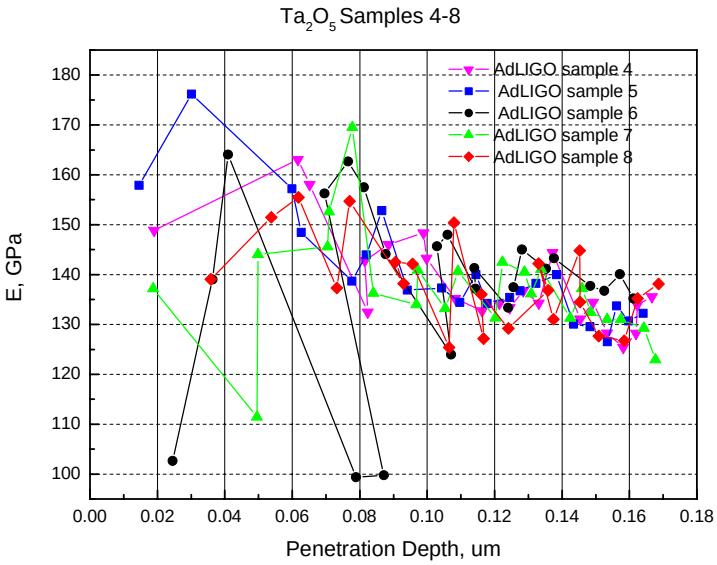

Fig. 3. Elastic modulus as a function of penetration depth for the films with different stoichiometry.

Typical thin film microhardness instruments can only produce sensible results if the penetration depth of the probe is no more than about 10% of the film thickness. For greater penetrations, the effects of the underlying layers or substrate become apparent. This can be seen from the general downward trend in modulus observed beyond about 80 nm depth in Fig. 2. In addition, penetration depths less than about 5% of the film thickness are prone to other errors, such as initial contact point (see Appendix). Thus the modulus for all the films is within 150±15 GPa at the 10% penetration depth (~70-80 nm).

The modulus as a function of penetration depth for a ~1 μm thick Ta_2O_5 film deposited with 45 sccm of oxygen backfill before and after annealing at 250°C, at 350°C and 450 °C is shown in Fig. 4. At the 10% penetration point, the modulus of all the films is within 140±10 GPa, which is basically the same as the result quoted above for oxygen level variation.

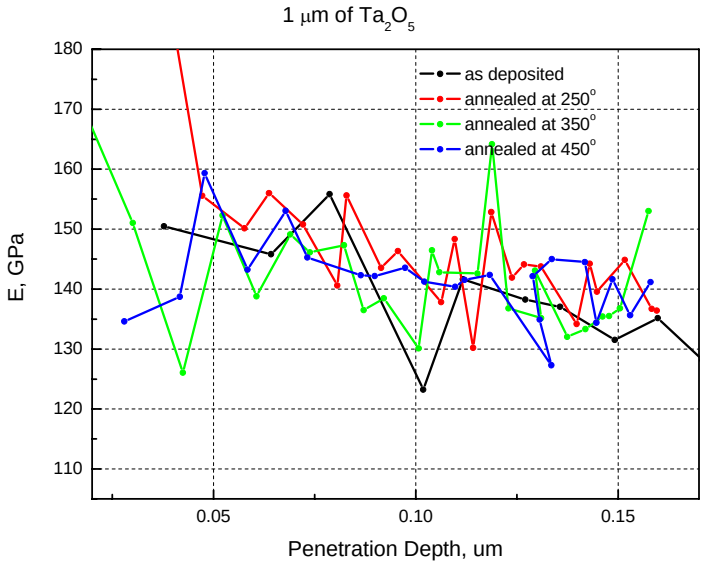

Fig. 4. Elastic modulus as a function of penetration depth for the 1 μm Ta_2O_5 film before and after annealing at 250°C, at 230°C and at 450°C.

Conclusions

- Varying the oxygen backfill level during an IBS deposition can vary the stoichiometry of the deposited tantala films.
- The absorption of a film with only a 10-20% increase in Ta increases by than 1000 times. The films are visibly highly absorbing and are of extremely poor optical quality, even at 1064 nm. A mirror created from such material would appear almost black at visible wavelengths.
- The changes in the stoichiometry achieved here do not result in measurable changes in Young's Modulus of the tantala films.
- Annealing of the tantala films in air at temperatures up to 450°C do not result in measurable changes in Young's Modulus.

Appendix

Ultra-micro Indentation Hardness Testing (UMIS)

Indentation experiments were carried out using Ultra-Micro Indentation System UMIS designed and constructed by CSIRO Division of Telecommunications and Industrial Physics. The UMIS website at www.tip.csiro.au/umis contains additional information about UMIS and the WinUMIS software.

Independent force and depth measurements were performed to obtain information on elastic properties of the deposited films. The structure of a typical test cycle is as follows. A test cycle consists of an initial contact (0.015 mN), followed by a loading step, where the load is incremented (24 increments of 0.2mN) up to a set maximum load (0.2mN for the first measurement). At maximum load, a hold time 0.1 sec was carried out, then unloading to 2% of maximum was performed in 20 increments. At the end of unloading, a further hold period 0.1 sec was implemented.

Measured data were stored as text files. (A set of measurements for one of the samples (AdLIGO 4) is attached as an example). Each test data file has an initial contact (coded as "IC") and a maximum load coded "Maximum". Maximum load occurs after any hold period. In the analysis, thermal drift can be computed from either the hold period at maximum load or unload. 24 indentations were performed in each test cycle with 50μm spacing between the indentation positions. Square root increments of the loading/unloading force sequence were used in order to take more measurements in the near-surface region, where property changes can be most dynamic. Delay till the starting time of a data acquisition sequence was set up to 30 minutes to insure thermal equilibrium of the system.

Raw measurement set of data for the test sample with 45 sccm of oxygen backfill is presented in Fig. 6 as an illustration.

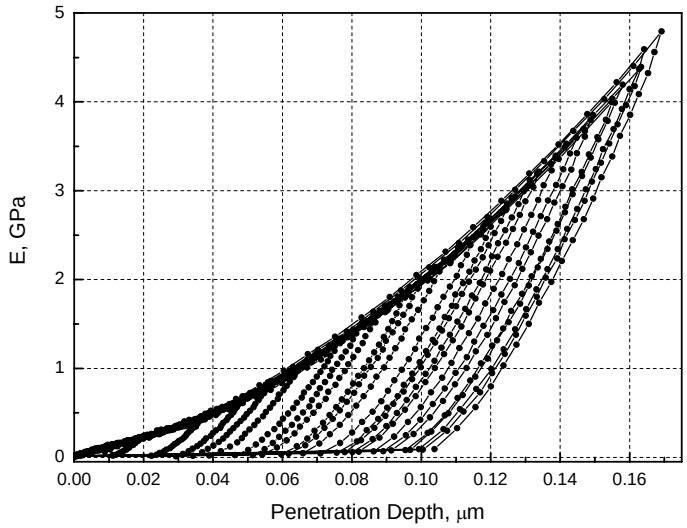

Fig.6 Typical set of measurements (45 cc oxygen backfill sample)

Raw data were corrected for the indenters' shape using a lookup table generated by a 'tip profiling' module as well as for instrument compliance and initial penetration.

An area function file contains ratio information to correct for the non-ideal shape of a real indenter. The data for an area function was created from a series of tests on a specimen of known modulus (fused silica).

To correct for the displacement arising from the deflection of the load frame of the instrument compliance was set at 0.000750μm/mN. The instrument compliance value was determined from a plot of dh/dP vs 1/hp

The indenter must make a small contact with the specimen surface before the depth measurements can be taken in an indentation test. An initial contact force was set at 0.015 mN and correction value for the initial penetration depth has been calculated by special logarithmic fitting routine (10 points). The initial penetration depth, hi, was added to all depth measurements, h, to correct for this initial displacement.

From the measured data elastic modulus has been calculated as a function of depth using theoretical models. The analysis the “multiple-point unload method” (developed by Oliver and Pharr) has been applied to data obtained with Berkovich indenter. The multiple-point unload method has been set up to use 7 data points on the unload part of the test and to fit 2nd order polynomial curve to these data. The slope of this curve at the force under consideration was then extrapolated to the x axis and various corrections applied to this intercept to find a value of the plastic depth hp. The slope of the line is

used to determine the combined modulus E* using the derivative of the Hertz elastic equations. For the multiple-point unload method, the tangent to the line of best fit at intermediate and final loads is used to determine the quantities of interest. The shape factor b (b=1.034) is a geometric correction to account for the non axis-symmetric nature of a real Berkovich indenter. The h intercept factor is “correction” factor applied to the intercept of the tangent to the curve with the x axis. A value of h=0.75 has been found to provide results which are more comparable to those obtained by other methods.

The overall results of interest are the combined elastic modulus E* and the hardness H. The hardness is the computed hardness of the specimen material as found from an estimation of the contact area (vie the “plastic depth hp) and the indenter load adjusted for the non-ideal shape of the indenter and other corrections. The combined modulus E* is presented as E* for the specimen and indenter combined, or E for the specimen only if a values for specimen and indenter Poisson’s ratio 0.23 and 0.07 respectively) and indenter modulus (1050GPa) is given in the analysis test parameters.

LIGO Contract No. 1063039
CSIRO Report 2
June 2004
Ion Beam Deposited Coatings

Objectives

- Test effect of ion bombardment assisted deposition on Young's Modulus of Tantala films.

Method

Tantalum oxide films were deposited by ion beam sputtering from a tantalum metal target. During the deposition the films were bombarded by an oxygen/argon ion beam from an assist ion gun. The deposition temperature was about 100ºC. Layers of tantalum oxide approx 700-800 nm thick were deposited on polished (commercial grade) homosil disks 25 mm diameter and 5 mm thick, with a fixed oxygen partial pressure in the deposition chamber. The sum of the oxygen flows from the assist ion source and the passive oxygen inlet was 50 sccm for all experiments. 1 sccm of Argon was admitted into the assist gun to provide discharge stability and easier ignition, but is not expected to add significantly to any ion bombardment effect.

The optical characteristics of the films were determined from transmission measurements in a Varian Cary 5 UV-Vis-NIR spectrophotometer. Woollam's *WVASE32* software was used to model the transmission characteristics to extract the refractive index, thickness and extinction coefficient.

The Young's Modulus of the films was measured using a CSIRO manufactured ultra-micro indentation system (UMIS). An explanation of this system and its limitations has been provided in Report 1.

Results

Optical characteristics

Three samples were deposited using different ion bombardment energies, but with a constant oxygen/argon beam current of 100 mA (approx 150 μA cm^{-2} at the substrate). The optical constants
of non-annealed films at 1064 nm are shown in Table 1.

Table 1

Beam Energy	0 eV	300 eV	600 eV	1200 eV
RI	2.11	2.09	2.09	2.08
K	.00004	0.00002	0.00003	0.00003

Rate (nm/sec)	0.275	0.288	0.282	0.281

There is some suggestion that the ion assist process increases the deposition rate slightly, although with increasing energy, etching of the film results in a decrease in this effect. The refractive index also decreases with ion bombardment, suggesting that oxygen is being implanted in the film at a greater than stoichiometric level. A purely Ar ion assisted deposition is expected to result in an increase in refractive index as the Ar can preferentially sputter oxygen out of the growing film leaving it sub-stoichiometric. This could be seen in the XPS results in Report 1 (Note: problems with the XPS instrument have prevented us from measuring the stoichiometry of these films at this time, however, these measurements will be available with the next report).

The extinction coefficients appear to benefit slightly from low energy ion bombardment, however, given the limited accuracy of loss measurements obtained from modelling, this may not be significant. In addition, all samples give a modelled k value of 0 after annealing at 350°C for 2 hours, suggesting that whatever the effects of ion bombardment during deposition, they are essentially nullified by annealing.

Young's Modulus

Figure 1 shows the elastic (Young's) modulus as a function of probe penetration depth for the various ion bombardment energies used above.

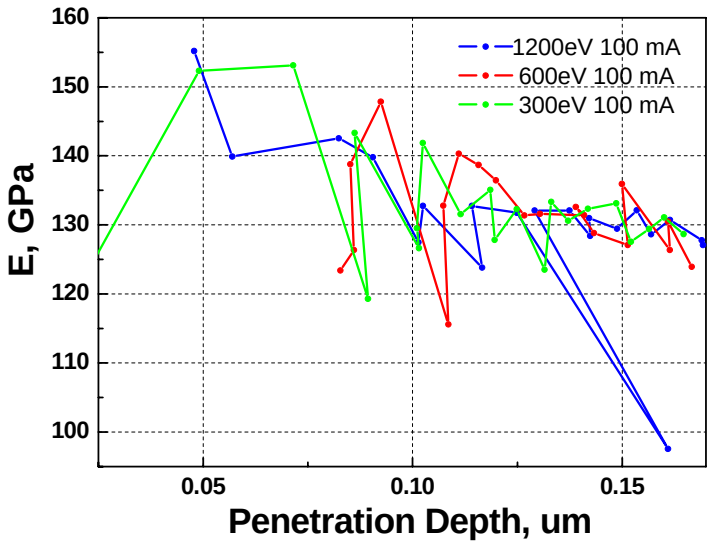

Fig. 1. Elastic modulus as a function of penetration depth for the films deposited with different ion bombardment energies.

The modulus for all the films is essentially indistinguishable at 140±15 GPa for penetration depths of 5-10% of the film thickness (0.04-0.08 μm). This is slightly lower than the value obtained for the unbombarded samples in Report 1 (approx 150 GPa) which may be explained by the possible implantation of additional oxygen suggested by

the optical results. Nevertheless, any change in Young's Modulus is certainly small. For comparison, the modulus of the silica substrate, measured using the same instrument, is 75±10 GPa.

Microcrystallinity

X-ray diffraction measurements of the above films have shown no evidence of possible ion-bombardment induced microcrystallinity, the as-deposited films being amorphous according to this test. Furthermore, annealing of the films at 600°C for 2 hours did not change this result. At 700°C, however, strong evidence of crystallization of the film was obtained, both from the x-ray spectra and visually since the films became cloudy in appearance. The grain size is unknown. This is in agreement with other sources which have shown the threshold of microcrystallization of an initially amorphous tantala film to be somewhere between 600°C and 700°C . The Young's Modulus of the microcrystallized films has been measured at approximately 130±25 GPa. The large uncertainty is due to the inhomogeneity of the film at the level of the probe tip size. Although the result is again slightly lower than the results obtained before, such films are of little value due to the very large scattering inherent in microcrystalline films.

Conclusions

- Ion bombardment assistance of ion beam sputtered tantala films produces a possibly slightly oxygen rich film according to optical measurements.
- The Young's Moduli of these tantala films is slightly lower than those of non-bombarded films, but not dependent on ion energy to any significant degree.
- There is no microcrystallinity in the films and this remains so even when they are annealed at temperatures up to 600°C.
- The Young's modulus of films heated to the point of microcystallization is the lowest value so far measured. This perhaps suggests a lower limit to the degree of manipulation of the Young's modulus of tantala using purely physical techniques (i.e. without gross chemical changes to film achieved by, for example, co-doping).

LIGO Contract No. 1063039
CSIRO Technical Report 3
July 2004
Ion Beam Deposited Coatings

Objectives

- To reconfigure the IBS system for planetary substrate motion and establish conditions for coating of sapphire flats and dimpled substrates.

Report

Until recently the set-up of the IBS system remained close to that as used to produce DWDM's by Spectra Physics. In that Spectra Physics arrangement the target-to-substrate distance was very short, and the area of good and reproducible uniformity was only over a few millimetres diameter annulus a considerable distance from the centre of rotation of the substrate. Some initial modifications to the arrangement were made to obtain reasonable uniformity over a somewhat greater substrate aperture. That is the set-up used in our initial experiments for the AdvLIGO contract. However, this was not suitable for the thicker sapphire flats, both from a dimensional point-of-view, as well as a reproducible uniformity over the distances between the substrate pairs and the witnesses. In the longer term these changes were needed to better replicate the operating conditions of the system for larger substrates.

The latest modifications have been involved moving back the whole substrate platform to substantially increase the target-to-substrate distance. This required a nipple to be added to the substrate flange, and for all the rotation and electrical feedthroughs to be rerouted. A planetary motion system (2 planets) has been installed to provide substrate fixturing. A considerable amount of time has been spent establishing the best conditions for single beam IBS with this configuration. Coating uniformity has been considerably improved, as has the reproducibility of the coating properties (refractive indices and layer thicknesses). The rates of deposition of the materials have decreased by a factor of about 60%, so coating duration is longer.

Initial results indicate that the spectral reproducibility of multilayer HR-coatings is better than 0.5% and the reproducibility of the minimum transmittance of annealed samples well within the specification for the HR for the present sapphire coating (650ppm, +50ppm, –0ppm). Coating of the HR on the dimpled surfaces begins tomorrow (29/7/04). Progress on optimising single and multilayer properties has been slower than anticipated because there is no optical monitoring in the IBS system. There are funds now available to build an ellipsometer for in-situ measurement and control during the financial year ending June 05. There is also funding to build a heavy-duty planetary motion system for substrates up to 350mm diameter and 40kg mass.

Plans for immediate future

(a) HR on dimpled substrates. Both in same run.

(b) Please ship the sapphire flat asap

(c) AR coating trial
(d) AR coating of flat surface of dimpled substrate
(e) HR on sapphire flats
(f) AR on sapphire flats
(g) HR with substoichiometry Ta_2O_5 and SiO_2 for Q testing
(h) HR on superpolished and commercially polished substrates for Q test comparison
(i) Resume development work

1 November 2004

Advanced LIGO coatings Ion Beam Sputtering

Several issues related to the performance of our ion beam sputtering system have hampered our progress on low mechanical loss coatings. This is a second-hand Veeco-IonTech Spector IBS system originally configured for DWDM applications in the photonics industry. We have been attempting to reconfigure this machine for more conventional coating requirements and have found that this has not been as straightforward as we had imagined. Some of the issues and our attempts to rectify them are as follows:

Coating Uniformity

Our original IBS system came configured for DWDM coating of a fixed substrate type, where uniformity was determined solely by fixed masks. One consequence of this design was that changing the substrate dimensions or positions always resulted in considerable trial-and-error effort to redesign these masks. We have now fabricated a simple planetary stage, which should mean that uniformities better than 1% can be achieved routinely on any LIGO test substrates. One issue still to be resolved is the contribution of the motion of this stage to particulate generation in the system (see below). We have some experimental evidence to suggest, that in some cases, the motion of the stage somehow results in particulates accumulating on the substrates under the edge of the aperture of masks that have been places over the substrates to limit the coating extent. This may be an electrostatic charging issue, but seems to be dependent on the aperture diameter. We have not resolved these problems to date. We have a plan in place to construct a more robust stage, which should give even better uniformity in the future.

Substrate Cleaning

One some of the previous LIGO coatings, it was reported that ‘cleaning marks’ were visible in the coated surfaces. We have now put in place a more robust cleaning regime that should see this problem reduced. However, we are still developing the full infrastructure for proper substrate cleaning and until this is complete, cleaning will continue to be problematic.

Particulates

Particulate contamination of varying severity is always present in the coatings. We have not been able to identify the origin of these particulates with any certainty, but they appear to be worse when the ion gun discharge is on (beam on or off), suggesting electrostatic attraction due to charging may be the main cause. While this is probably a problem with all IBS systems, we have no data from other systems with which to compare our results. The LIGO I and II specifications give figures for total scatter loss, but do not distinguish between scatter from the bulk of the film and from the particulates (eg. a ‘defects per square centimetre’ specification). Nevertheless, some work on other

projects has suggested that our particulate levels are higher than one might expect and hence our ongoing effort to reduce them.

Main system cryogenic vacuum pump

This, it now appears, has been a problem since we purchased our IBS system. The symptom has been overheating of the pump partway through a coating run, necessitating a shutdown until the pump cools down, and the requirement for frequent regenerations. Excess gas flow and chamber heating were suggested by Veeco as causes, as well as the fact that we run a mains electrical frequency of 50 Hz in Australia, whereas the pump is designed for 60 Hz. However, now that the pump has been replaced with a new one, it is clear that the pump was at fault and had been slowly degrading, until a sudden and obvious decline in performance occurred several months ago, prompting its replacement. Uninterrupted and reliable coating runs should now be possible, which should increase throughput.

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Wednesday 8, December 2004

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Progress report for LIGO Contract No. 1063039 – Dec 2004

Work since the last report in July 2004 has been primarily directed at:

- Final coating runs and measurements of four sapphire test masses (70 mm D x 60 mm thick cylinders) and samples for Prof. Braginsky.
- Progress on upgrading planetary motion stage for 150 mm diameter substrates (Option A of Start up and infrastructure preparation cost of contract). This work is ~ 33% complete.
- Coated and delivered three 3"x 0.1" standard LIGO test substrates with 30 layer TaO/SiO coatings for tests of mechanical loss dependence on tantala layer stoichiometry and commercial versus superpolished substrates.
- Performed trial runs to determine IBS machine conditions for operation with Xenon (instead of Argon) sputter gas.
- Performed trial coating runs using Xenon process conditions to determine layer deposition times for 30 layer mirrors.
- Fine tuned fixturing to run thick and thin LIGO test substrates in same IBS run with Xenon process.

A separate technical report *("Technical Report 4 – Dec 2004.doc")* on the results achieved since the last report will be submitted shortly.