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Tantalum Oxide and Silica Mixture Coatings Deposited Using Microwave Plasma Assisted Co-sputtering for Optical Mirror Coatings in Gravitational Wave Detectors

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Abstract: This work presents the characterisation of optical and mechanical properties of thin films based on $(Ta_2O_5)_{1-x}(SiO_2)_x$ mixed oxides deposited by microwave plasma assisted co-sputtering to demonstrate their potential as optical coatings in gravitational wave detectors.

OCIS Codes: (310.1860) Deposition and fabrication; (310.6845) Thin film devices and applications; (310.3840) Materials and process characterization

1. Introduction

Gravitational-wave detectors (GWDs) such as Advanced LIGO (aLIGO), Advanced Virgo (AdvVirgo) and KAGRA are ground-based interferometric detectors utilising high finesse optical cavities to perform high precision displacement measurements. The test mirrors of these GWDs consist of a high purity silica (SiO_2) substrate coated with a high reflecting (HR) coating, Bragg's reflector alternating layers of low and high refractive-index amorphous materials typically deposited by ion-beam sputtering (IBS). LIGO and Virgo originally employed SiO_2 and Ta_2O_5 as the low (n = 1.46) and high (n = 2.12) index materials, respectively. It is worth noting that for the mirror coating used by LIGO and Virgo, the major source of thermal noise was Ta_2O_5, hindering overall sensitivity of the GWD [1]. The low index material (i.e. SiO_2) exhibited a minor contribution to this problem. To address this issue, aLIGO and AdvVirgo upgraded the mirror coatings to use TiO_2:Ta_2O_5 mixture for the high index material [2], reducing mechanical losses (ϕ) by around 40% and increased the refractive index up to n = 2.19 [3]. This promising result leads to new investigations and explorations for other oxide mixtures of high index that might feature low mechanical loss while preserving the high index or even augmenting it [4].

Investigations for various random network oxides showed that the annealed SiO_2 and GeO_2 mixed with Ta_2O_5 featured the lowest loss angles, however, the Brownian thermal noise considers not only the loss angle of the material but also the total thickness of Bragg's reflector stack. Thus, ideal properties of the high index material comprise of low ϕ and high n at λ = 1064 nm. Unfortunately, it has been observed that a decrease in the ϕ leads to a decrease in the n of the material. For this reason, we investigated new deposition techniques alternative to IBS with aims to deposit low ϕ materials while maintaining/increasing n.

This work presents a study of optical and mechanical properties of tantala/silica $(Ta_2O_5)_{1-x}(SiO_2)_x$ mixture coatings deposited by microwave plasma assisted co-sputtering technique with low optical absorption and low mechanical loss for gravitational wave detection. This deposition technique presents advantages with respect to standard IBS, including higher precise deposition control, higher deposition rate and better consistency. Further investigation on thermal treatments will follow.

2. Deposition Process and Characterization Methods

Deposition was carried out using a microwave plasma assisted pulsed DC reactive sputtering process. Within the deposition system there is a horizontal axis rotating drum in which deposition of each oxide layer can be achieved with multiple passes through rectangular planar DC magnetron sources and assisted microwave plasma oxidation region. The high deposition rate is obtained through the metal-like sputtering and a separated microwave plasma assisted oxidation reactive deposition which avoids target poisoning [6, 7]. To produce the mixtures of Ta_2O_5 and SiO_2, two targets, Ta and Si, were mounted on left and right sides of chamber and sputtered simultaneously. Quartz crystals are used for thickness monitoring and calibrated for both targets separately prior to co-sputtering. The composition of mixtures was adjusted by varying the sputtering power of Ta target. Detailed process parameters are shown in Table 1. Samples 1 and 2 are used to calibrate crystal monitoring. For mixtures, the deposition rate is the exactly the individual layer thickness, as the drum rotates at 60 rpm, i.e. one revolution per second, thus individual layer thickness is in the range of Angstroms.

For optical property analysis, samples were deposited on JGS3 substrates (SiO_2 films have a layer of Ta_2O_5 deposited prior to SiO_2 deposition due to similar n of SiO_2 and JGS3). n and k were obtained by fitting the transmission data obtained by a Perkin-Elmer Lambda 40 spectrometer and employing multiple Kim's oscillation model [8].

Table 1. Sample list and parameters of $(Ta_2O_5)_{1-x}(SiO_2)_x$ mixed oxide films

Sample No	Ta target, power control			Si target, voltage control			Expected Ta_2O_5 Volume Fraction
	Power (KW)	Deposition Rate (Å /s)	Expected Thickness (nm)	Voltage (V)	Deposition Rate (Å /s)	Expected Thickness (nm)	
1	3.5	2.7	500	0	0	0	1
2	0	0	0	400	0.85	500	0
3	3.5	2.7	380	400	0.85	120	0.76
4	2.4	1.9	345	400	0.85	155	0.69
5	1.5	1.1	256	400	0.85	244	0.51
6	0.9	0.6	207	400	0.85	293	0.41

The Q factor measurements are performed by exciting a resonant mode of the sample and measuring the exponential decrease in the oscillation amplitude (ringdown). The coatings were deposited on pre-annealed (1000°C for 4 hours) fused silica substrates of diameter of 76.2 mm, thickness of 511±1μm and a flat of 25 mm. To test the gentle nodal suspension (GeNS), the cylindrical shaped sample is suspended on a silicon flat-convex lens with lens positioned over an aluminium platform and firmly held by a conical hole [5]. This GeNS is hosted inside a vacuum tank (<2×10^{-6} mbar), with He-Ne laser (λ = 633 nm and 4 mW) and a quadrant photodiode to measure the displacement of the sample excited at different resonant frequencies. Ringdown measurements record the decay of the excited resonant mode amplitude of the sample exhibiting damped harmonic motion, consisting of carrier and envelop signals, where the amplitude of the envelope can be represented by:

$$A(t) = A(0)\exp[-\pi f_0 \phi(f_0) t] \quad (1)$$

The mechanical loss of the coating (ϕ_{coating} coating) is calculated as follows:

$$\phi(\omega_0)_{coating} = \frac{E_s}{E_c}[\phi(\omega_0)_{coated} - \phi(\omega_0)_{substrate}] \quad (2)$$

Where E_s/E_c is the substrate/coating energy ratio estimated using finite element analysis (ANSYS).

3. Optical Properties of the Co-sputtered films of Tantalum Oxide and Silica

Figure 1 shows the *n* and *k* represented as a function of wavelength (λ). The fitting method used to calculate *n* resulted in highly accurate values, particularly at a λ of 1064 nm, for both pure Ta_2O_5 (1) and SiO_2 (2) samples. However, the transmittance measurement accuracy of the spectrophotometer limits the extinction coefficient accuracy for low absorption materials. From Figure 1, one could conclude that *n* of the resulting mixture decreases with Ta_2O_5 volume fraction, and the absorption of mixture decreases when Ta_2O_5 is mixed with SiO_2, particularly for the 300 nm absorption edge of Ta_2O_5/SiO_2 mixture (*see* Figure 1 insert).

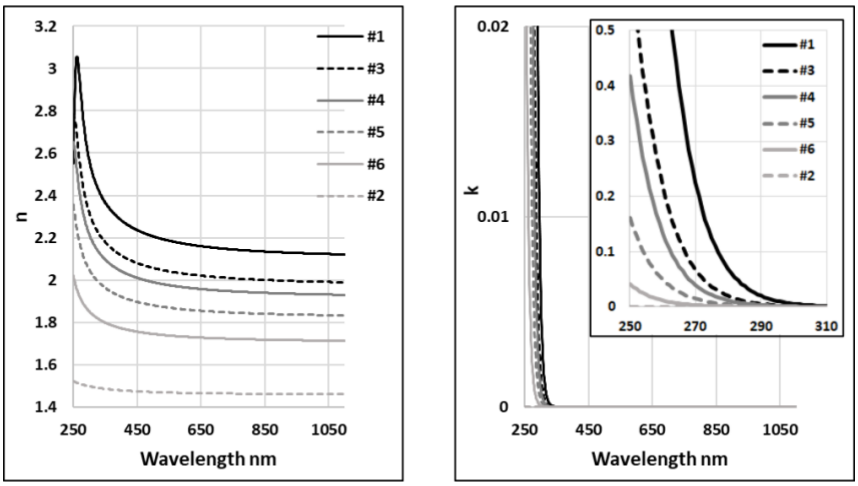

Figure 1. Wavelength dependence of the *n* and *k* for $(Ta_2O_5)_{1-x}(SiO_2)_x$ co-sputtered coatings.

Based on optical properties of the mixture, the volume ratio can be obtained by using a generalized anisotropic Bruggeman Effective Medium Approximation (EMA) [9]. The actual coatings are the stacks of alternative layers of tantalum oxide and silica with thickness in the angstrom range. As there are only two phases in our samples, this EMA equation can be simplified to:

$$f_1\frac{\varepsilon_1-\varepsilon}{\varepsilon_1+2\varepsilon} + (1-f_1)\frac{\varepsilon_2-\varepsilon}{\varepsilon_2+2\varepsilon} = 0 \quad (3)$$

Where the dielectric constants for phase 1 (Ta_2O_5, ε_1), phase 2 (SiO_2, ε_2) and the mixture (ε) are known. Volume fraction of Ta_2O_5 (f_1) can then be calculated using Eq(3) as shown in Table 2. Discrepancy between calculated and expected volume fractions may be from deposition rate reading accuracy as the reading for deposition rate on IC5 is angstrom level and only has 2 digits.

Table 2. Calculated volume fraction of Ta_2O_5 using Eq(3) and the refractive index at λ = 1064 nm.

Sample No	n	ε	Expected volume fraction of Ta_2O_5	Calculated volume fraction of Ta_2O_5	Expected thickness (nm)	Fitted thickness (nm)
1	2.122	4.503	1	1	500	486.9
2	1.460	2.132	0	0	500	533.0
3	1.990	3.960	0.76	0.806	500	492.4
4	1.930	3.725	0.69	0.719	500	487.8
5	1.833	3.360	0.51	0.577	500	467.7
6	1.713	2.934	0.41	0.399	500	502.0

4. Mechanical Loss of the Co-sputtered films of Tantalum Oxide and Silica

The resulting mechanical loss angles, including substrate ($\phi_{\text{substrate}}$), coated (ϕ_{coated}) and coating (ϕ_{coating}) loss for all materials are summarized in Table 3. The table also includes the substrate/coating energy ratio (E_s/E_c) extracted for the first fundamental resonant mode using ANSYS and used here to calculate ϕ_{coating} using Eq(2).

Table 3. $\phi_{\text{substrate}}$, ϕ_{coated} and ϕ_{coating} of single layer coatings measured at the first resonant mode.

Material (*thickness*)	**Frequency** (Hz)	$\boldsymbol{\phi_{\text{substrate}}}$ ($\times10^{-6}$)	$\boldsymbol{\phi_{\text{coated}}}$ ($\times10^{-6}$)	$\boldsymbol{E_s/E_c}$	$\boldsymbol{\phi_{\text{coating}}}$ ($\times10^{-4}$)
Ta_2O_5 (*500 nm*)	530.62	1.82 ± 0.01	12±2	148.04	14±4
Ta_2O_5 (80%) / SiO_2 (20%) (*500 nm*)	527.95	4.11 ± 0.06	10.9±0.4	147.71	10.3±0.5
Ta_2O_5 (75%) / SiO_2 (25%) (*500 nm*)	532.96	1.71 ± 0.01	5.2±0.5	148.74	5.7±0.7
Ta_2O_5 (62.4%) / SiO_2 (37.6%) (*500 nm*)	528.53	1.35 ± 0.01	5.1±0.4	147.46	5.9±0.6
Ta_2O_5 (42.8%) / SiO_2 (57.2%) (*500 nm*)	532.61	2.52 ± 0.01	6.2±0.6	148.56	5.7±0.9
SiO_2 (*530 nm*)	521.43	3.83 ± 0.02	9.6±0.6	246.14	15±2

Results show that coating loss of mixed oxide films are in the range of 10^{-3}-10^{-4}, were a SiO_2 adding of around 57.2% resulting in 5.7±0.9 $\times10^{-4}$ without annealing treatment. These results are promising since the application of a thermal annealing is expected to reduce further the ϕ_{coating}, and could improve upon the current record values achieved in h-index materials consisting of TiO_2:Ta_2O_5, ranging between 2 and 3 10^{-4} [3].

5. Conclusions

In this work, we demonstrate a new deposition technique alternative to IBS allowing deposition of low mechanical loss materials (10^{-4}) with high refractive index (1.99). Our results for tantala/silica $(Ta_2O_5)_{1-x}$ $(SiO_2)_x$ mixture coatings deposited by microwave plasma assisted co-sputtering technique offer advantages with respect to standard IBS, including higher precision deposition control, higher deposition rate and better consistency.

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