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To cite this article: Alex Amato *et al* 2018 *J. Phys.: Conf. Ser.* **957** 012006

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High-Reflection Coatings for Gravitational-Wave Detectors: *State of The Art and Future Developments*

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Abstract. We report on the optical, mechanical and structural characterization of the sputtered coating materials of Advanced LIGO, Advanced Virgo and KAGRA gravitational-waves detectors. We present the latest results of our research program aiming at decreasing coating thermal noise through doping, optimization of deposition parameters and post-deposition annealing. Finally, we propose sputtered Si_3N_4 as a candidate material for the mirrors of future detectors.

The high-reflecting (HR) coatings of the gravitational-wave (GW) detectors Advanced LIGO [1], Advanced Virgo [2] and KAGRA [3] have been deposited by the Laboratoire des Matériaux Avancés (LMA) in Lyon (Fr), where they have been the object of an extensive campaign of optical and mechanical characterization. In parallel, an intense research program is currently ongoing at the LMA, aiming at the development of low-thermal-noise optical coatings. The materials presented in this study are deposited by ion beam sputtering (IBS), using different coaters: a commercially available Veeco SPECTOR and the custom-developed DIBS and Grand Coater (GC). Unless specified otherwise, each coater uses different sets of parameters for the ion beam sources. Coating refractive index and thickness are measured by transmission spectrophotometry at LMA using fused silica substrates ($\varnothing$ 1", 6 mm thick) and by reflection spectroscopic ellipsometry [4] at the OPTMATLAB using silicon substrates ($\varnothing$ 2", 1 mm thick). Results of the two techniques are in agreement within 3% and here are presented the average values, used to calculate coating density. Structural properties are probed by Raman scattering at the Institut Lumière Matière (ILM), using fused silica substrates. Finally, coating loss angle ϕ_c is measured on a Gentle Nodal Suspension [5, 6] (GeNS) system at LMA, with disk-shaped resonators of fused-silica ($\varnothing$ 2" and 3" with flats, 1 mm thick) and of silicon ($\varnothing$ 3", 0.5 mm thick). ϕ_c is evaluated using the resonant method [7] i.e. by measuring the ring-down time of several vibrational modes of each sample. For the i-th mode, it writes

$$\phi_{i,c} = \frac{1}{D_i} \left[\phi_{i,\text{tot}} - \phi_{i,s}(1 - D_i) \right] , \qquad D_i = 1 - \left(\frac{f_{i,s}}{f_{i,\text{tot}}} \right)^2 \frac{m_s}{m_\text{tot}} ,$$

where $\phi_{i,\text{tot}}$ is the loss angle of coated disk and $\phi_{i,s}$ is the loss angle of the substrate. D_i is

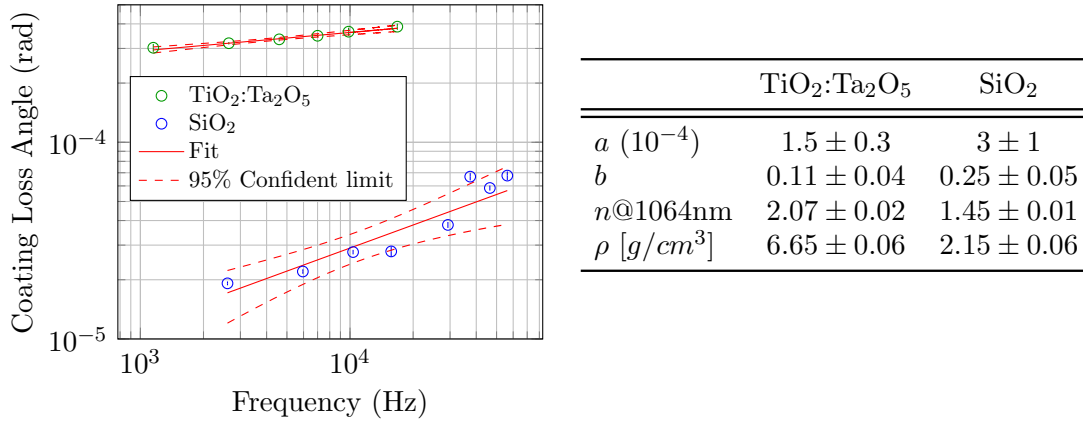|	TiO$_2$:Ta$_2$O$_5$	SiO$_2$
a (10^{-4})	1.5 ± 0.3	3 ± 1
b	0.11 ± 0.04	0.25 ± 0.05
n@1064nm	2.07 ± 0.02	1.45 ± 0.01
ρ $[g/cm^3]$	6.65 ± 0.06	2.15 ± 0.06

Figure 1: (Color online) Coating loss, fit parameters and optical properties of standard materials deposited by GC and annealed at 500℃ for 10 hours.

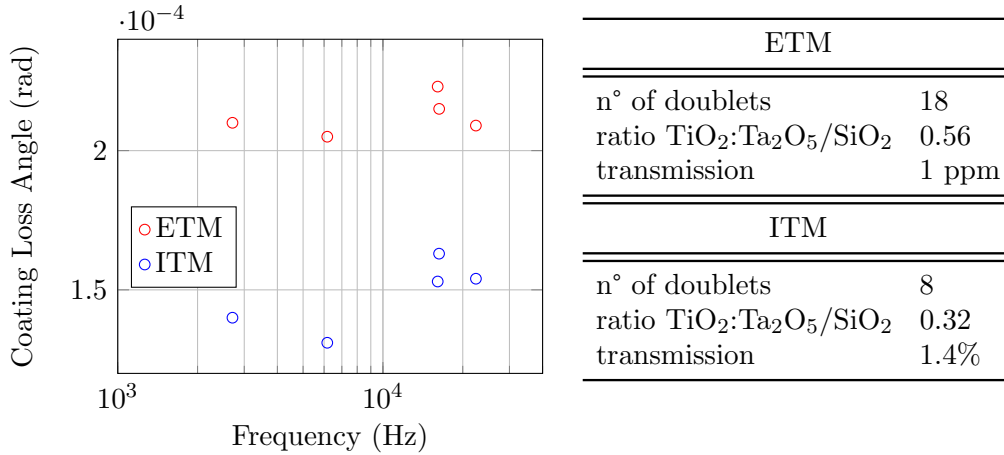| ETM	
n° of doublets	18
ratio TiO$_2$:Ta$_2$O$_5$/SiO$_2$	0.56
transmission	1 ppm
ITM	
n° of doublets	8
ratio TiO$_2$:Ta$_2$O$_5$/SiO$_2$	0.32
transmission	1.4%

Figure 2: (Color online) Coating loss and stack properties of ITM and ETM HR mirrors deposited by GC and annealed at 500℃ for 10 hours [6].

the so-called *dilution factor* which can be related to $f_{i,s}$, $f_{i,\text{tot}}$, m_s and m_{tot} [8], that are the frequencies and the mass of the sample before and after the coating deposition, respectively.

1. Standard materials in gravitational-wave interferometers
HR coatings of Advanced LIGO and Advanced Virgo are Bragg reflectors of alternate titania-doped tantala (TiO$_2$:Ta$_2$O$_5$) and silica (SiO$_2$) layers [1, 2]. Fig. 1 shows the mechanical loss of these materials, which seems to follow a power-law function $\phi_c = a \cdot f^b$. The loss angles of the HR coatings are shown on Fig. 2, together with their properties. The end mirror (ETM) coating has higher loss angle than the input mirror (ITM) coating because of its higher ratio TiO$_2$:Ta$_2$O$_5$/SiO$_2$.

2. Optimization
2.1. Doping
The purpose of TiO$_2$ doping is to increase Ta$_2$O$_5$ refractive index and reduce its loss angle. Increasing the refractive index contrast in the HR stack would allow to decrease the HR coating thickness, at constant reflectivity. Fig. 3a shows Ta$_2$O$_5$ coating loss as function of doping. The
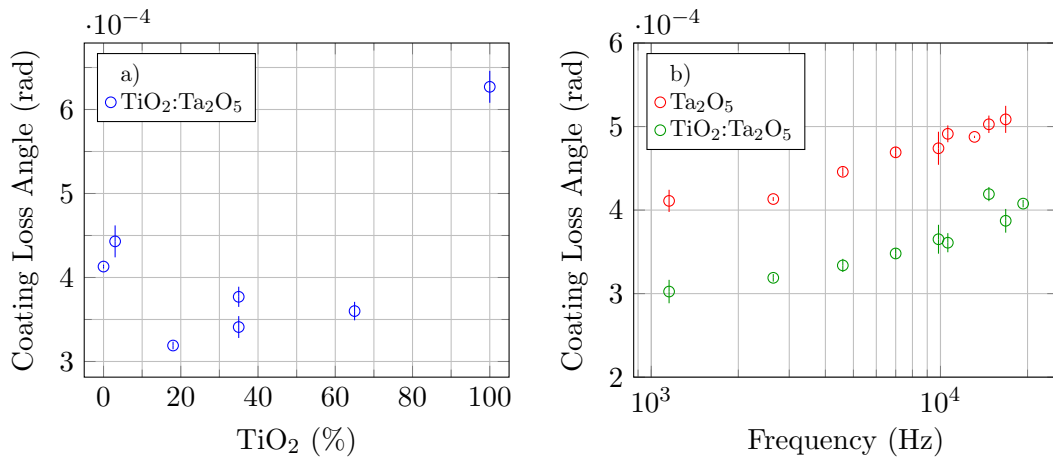
Figure 3: (Color online) a) Coating loss of TiO_2:Ta_2O_5 as function of TiO_2 doping. b) Comparison of Ta_2O_5 and 18%-doped TiO_2:Ta_2O_5 coating loss. All GC samples annealed at 500℃ for 10 hours (100% TiO_2 coating is crystallized).
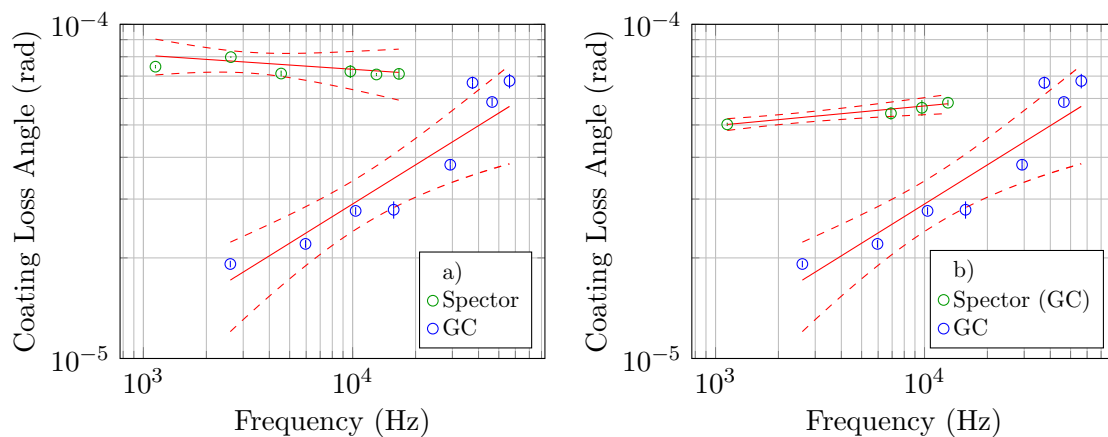

Figure 4: (Color online) Coating loss of SiO_2 deposited in the GC and in the Spector: a) different deposition parameters. b) same GC deposition parameters. All samples annealed at 500℃ for 10 hours. Fit model is the same as in Fig. 1.

current doping value in GW detectors is 18%, which yelds a minimum loss but a refractive index only slightly higher than that of Ta_2O_5. As shown by Fig. 3b, 18%-doped $\phi_{TiO_2:Ta_2O_5}$ is lower than $\phi_{Ta_2O_5}$ by ∼25%. Increasing TiO_2 concentration will increase TiO_2:Ta_2O_5 refractive index, while $\phi_{TiO_2:Ta_2O_5}$ for TiO_2 ≥40% can not be predicted and needs further investigation.

2.2. Deposition parameters

Fig. 4a shows coating loss of SiO_2 deposited by GC and Spector using their respective standard deposition parameters. It is clear that by using different parameters the same material gets different properties: GC parameters yield lower coating loss. For further test, SiO_2 has been deposited in the Spector with GC parameters. As Fig. 4b shows, Spector coating loss is lower but still higher than GC coating loss, because of the different configuration of the coaters. Coating losses of Ta_2O_5 deposited using different coaters have different values before annealing but converge toward a common limit value after, as shown in Fig. 7, suggesting that annealing

| | SiO$_2$ | | | Ta$_2$O$_5$ | |
	Spector	Spector as GC	GC	Spector	DIBS
a (10^{-4})	1.1 ± 0.2	0.33 ± 0.01	1.89 ± 0.09	2.6 ± 0.5	2.08 ± 0.08
b (10^{-1})	-0.4 ± 0.3	0.59 ± 0.05	1.00 ± 0.05	0.67 ± 0.08	0.88 ± 0.04
n	1.474 ± 0.005	1.468 ± 0.005	2.03 ± 0.02	2.08 ± 0.02	2.014 ± 0.005
ρ [g/cm^3]	2.34 ± 0.01	2.33 ± 0.05	7.34 ± 0.07	7.5 ± 0.1	6.9 ± 0.2

Table 1: Coating loss, fit parameters ($\phi_c = a \cdot f^b$), refractive index n at $\lambda = 1064$ nm and density ρ of materials deposited in different coaters.
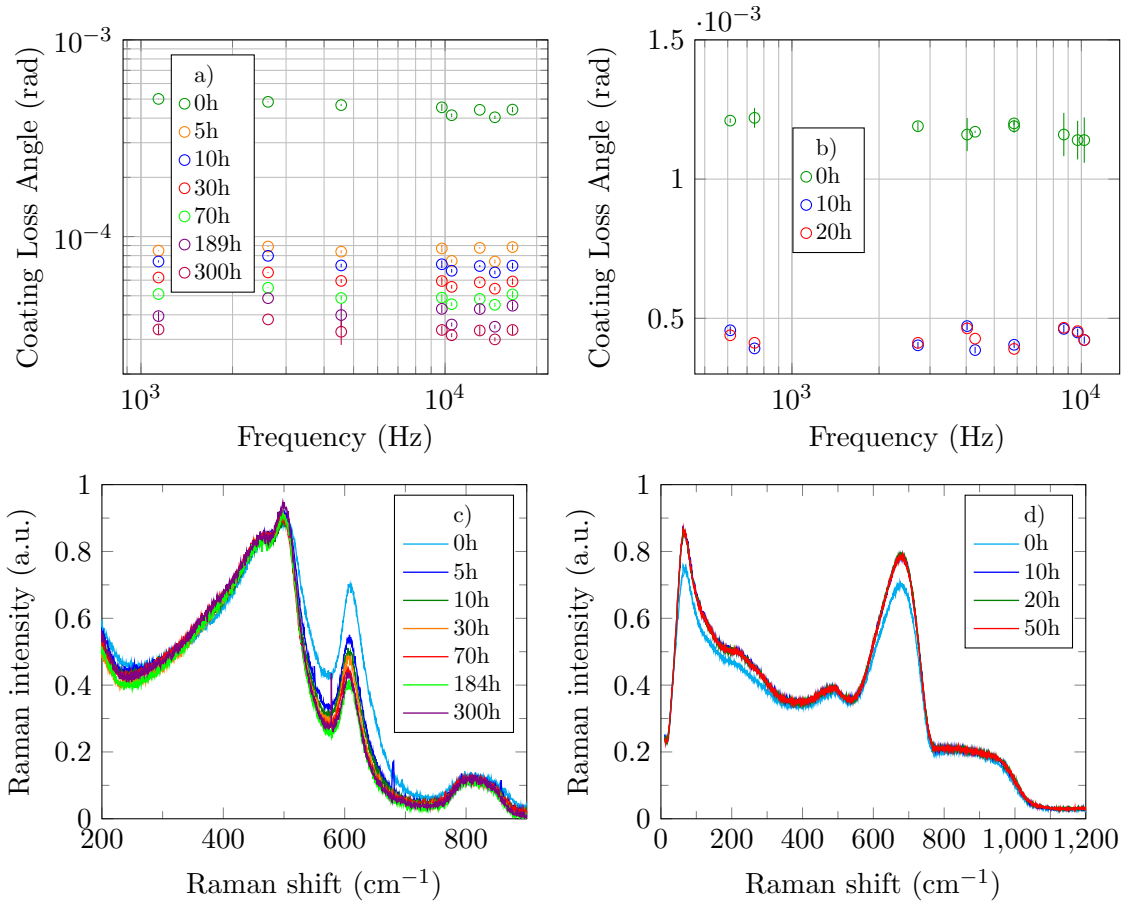

Figure 5: (Color online) Effect of annealing duration Δt [9]. top row: a) loss of SiO$_2$, b) loss of Ta$_2$O$_5$; bottom row: c) structure of SiO$_2$, d) structure of Ta$_2$O$_5$.

'deletes' the deposition history of the sample. Material properties are listed in table 1.

2.3. Post-deposition annealing

Annealing parameters are of fundamental importance for the purpose of reducing coating thermal noise. The problem is to find the optimal annealing temperature T_a and duration Δt, avoiding coating crystallization which would increase optical loss by scattering and absorption. In Fig. 5 is shown the effect of increasing Δt with $T_a = 500$°C constant. SiO$_2$ loss decreases and this behaviour has a structural counterpart. SiO$_2$ is composed of tetrahedral units arranged in
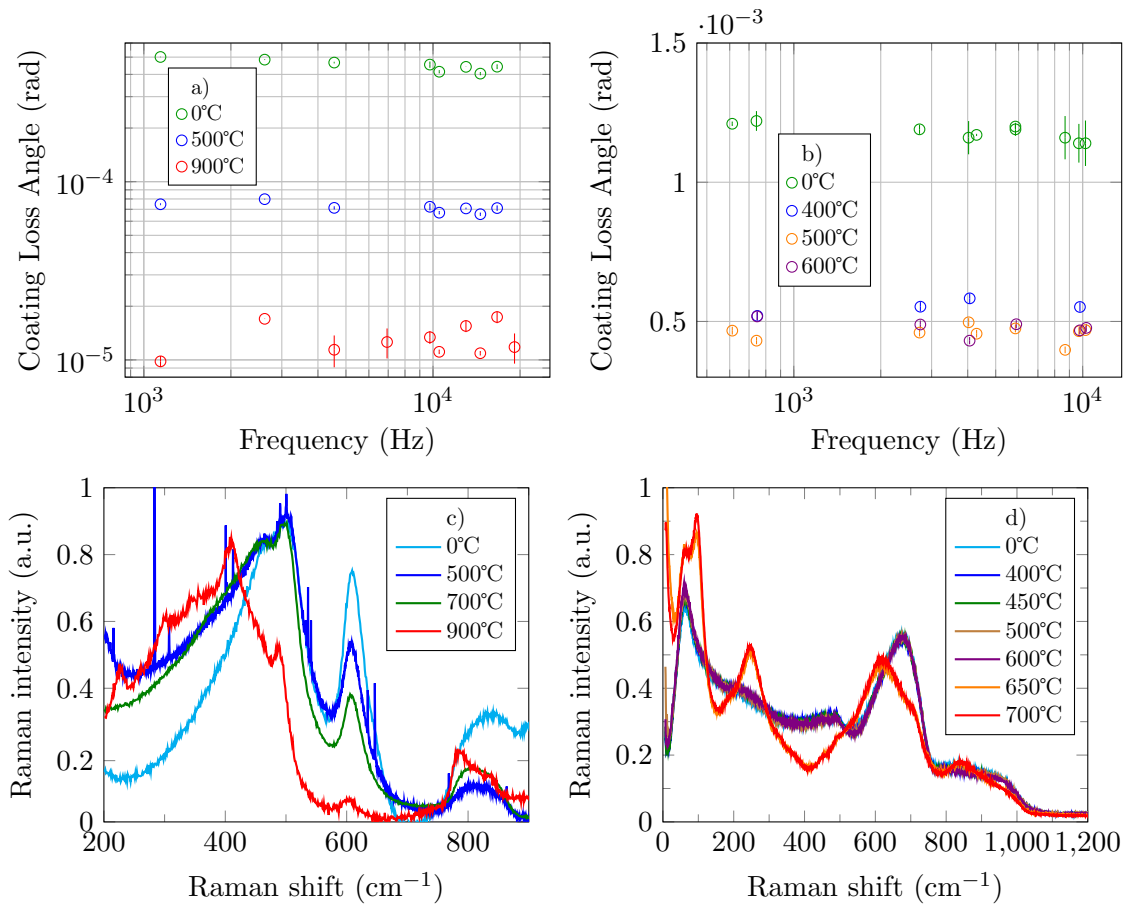
Figure 6: (Color online) Effect of annealing temperature T_a. top row: a) loss of SiO_2, b) loss of Ta_2O_5; bottom row: c) structure of SiO_2, d) structure of Ta_2O_5.
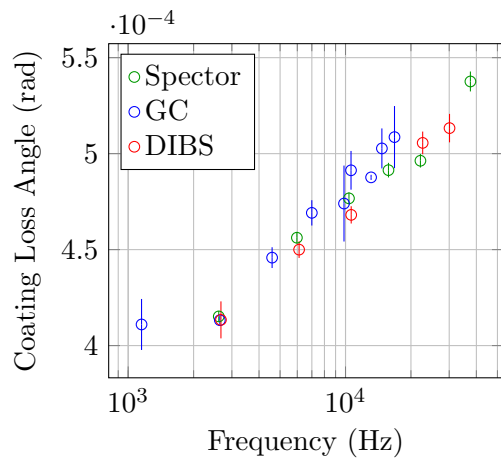

Figure 7: (Color online) Loss of Ta_2O_5 coatings deposited by different coaters and annealed at 500℃ for 10h.
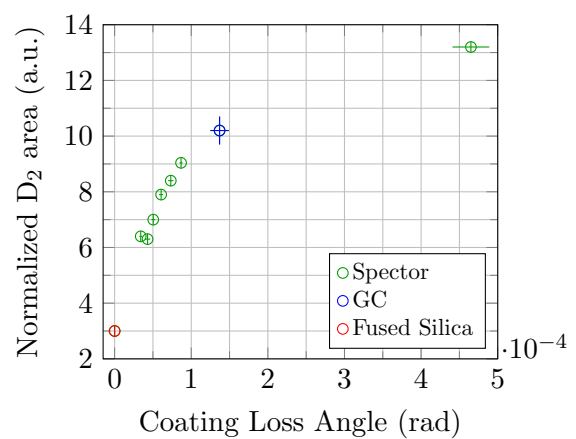

Figure 8: (Color online) Correlation between D_2 area and loss in SiO_2.

rings of different size [10] and the area of the D_2 band near 600 cm^{-1} is associated to 3-fold

ring population [11]. A correlation between coating loss and D_2 has been found, suggesting that SiO_2 loss increases with the 3-fold ring population [9]. This correlation holds for different kinds of SiO_2, coating and bulk (Fig. 8). On the other hand, Ta_2O_5 loss does not change for $\Delta t \geq 10h$ and its structure evolves only for $\Delta t \leq 10h$. Fig. 6 shows coating loss and structure for increasing T_a, with $\Delta t = 10h$ constant. SiO_2 loss decreases and its structure evolves considerably. Surprisingly, crystallization occurs at $T_a = 1000$℃. For Ta_2O_5, coating loss is roughly constant for $T_a > 500$℃ and its structure does not change up to $T_a = 600$℃, when crystallization occurs.
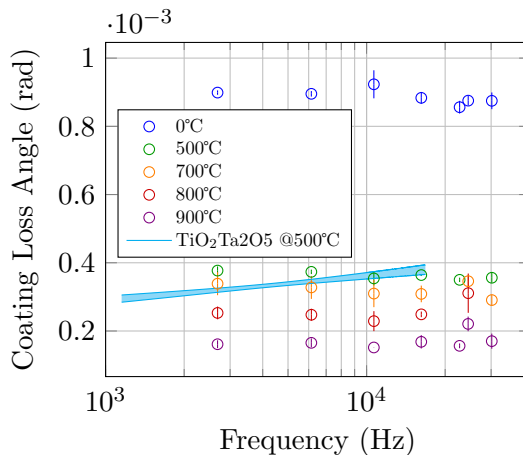

Figure 9: (Color online) Si_3N_4 loss for different T_a, compared to loss of TiO_2:Ta_2O_5 annealed at $T_a = 500$℃.

3. New material

TiO_2:Ta_2O_5 could be replaced by a material with lower mechanical loss and possibly higher refractive index. Here silicon nitride (Si_3N_4) is proposed, which features high refractive index [12] and very low mechanical loss [13]. Usually, Si_3N_4 is deposited by low pressure chemical vapour deposition (LPCVD). However, LPCVD Si_3N_4 might suffer from hydrogen contamination and thickness uniformity issues, which are not compatible with the stringent optical specifications required for GW detectors. Instead, IBS Si_3N_4 can be developed in the GC for deposition on large optics. Fig. 9 shows a comparison between TiO_2:Ta_2O_5 and IBS Si_3N_4, this latter being annealed at different temperatures. Si_3N_4 loss decreases significantly at $T_a = 900$℃. Thus, one could increase the annealing temperature of the entire HR stack to decrease also SiO_2 loss, eventually reducing the coating loss of the whole HR stack.

4. Conclusions

Coating materials of all present GW-detectors have been extensively characterized, showing a frequency-dependent loss angle. These standard materials can be optimized in different ways. The first approach is to increase the TiO_2 content in TiO_2:Ta_2O_5. Another option is to work on deposition parameters, in order to tune the optical and mechanical properties of the materials. In particular, the current GC configuration seems particularly well suited to deposit low loss SiO_2. In the case of Ta_2O_5, the effect of different deposition parameters is erased by the first few hours of post-deposition annealing. For 400℃$< T_a <$600℃ and 10h$< \Delta t <$50h Ta_2O_5 loss shows null or limited evolution and its structure is frozen in a stable configuration. In the case of SiO_2, the annealing parameters T_a and Δt have a significant impact on mechanical loss and coating structure. A correlation is found between D_2 peak area in the Raman spectra, associated to the three fold ring population, and mechanical loss. IBS Si_3N_4 is an interesting new possibility to replace TiO_2:Ta_2O_5 because of its low mechanical loss. Furthermore, Si_3N_4 can be annealed at higher temperature than TiO_2:Ta_2O_5, reducing also SiO_2 coating loss angle and thus the loss of the whole HR coating.

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