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 Si₃N₄ 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 (∅ 1”, 6 mm thick) and by reflection spectroscopic ellipsometry [4] at the OPTMATLAB using silicon substrates (∅ 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 Lumiére Matière (ILM), using fused silica substrates. Finally, coating loss angle $\phi_c$ is measured on a Gentle Nodal Suspension [5, 6] (GeNS) system at LMA, with disk-shaped resonators of fused-silica (∅ 2” and 3” with flats, 1 mm thick) and of silicon (∅ 3”, 0.5 mm thick). $\phi_c$ 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

$$\phi_{i,c} = \frac{1}{D_i} [\phi_{i,tot} - \phi_{i,s}(1 - D_i)]$$

where $\phi_{i,tot}$ is the loss angle of coated disk and $\phi_{i,s}$ is the loss angle of the substrate. $D_i$ is...
Figure 1: (Color online) Coating loss, fit parameters and optical properties of standard materials deposited by GC and annealed at 500°C for 10 hours.

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

the so-called dilution factor which can be related to \( f_{i,s}, f_{i,\text{tot}}, m_s \) and \( m_{\text{tot}} \) [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
current doping value in GW detectors is 18%, which yields a minimum loss but a refractive index only slightly higher than that of Ta$_2$O$_5$. As shown by Fig. 3b, 18%-doped $\phi_{\text{TiO}_2:\text{Ta}_2\text{O}_5}$ is lower than $\phi_{\text{Ta}_2\text{O}_5}$ by $\sim$25%. Increasing TiO$_2$ concentration will increase TiO$_2$:Ta$_2$O$_5$ refractive index, while $\phi_{\text{TiO}_2:\text{Ta}_2\text{O}_5}$ for TiO$_2 \geq 40\%$ cannot 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$_2$O$_5$ 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

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Figure 3: (Color online) a) Coating loss of TiO$_2$:Ta$_2$O$_5$ as function of TiO$_2$ doping. b) Comparison of Ta$_2$O$_5$ and 18%-doped TiO$_2$:Ta$_2$O$_5$ coating loss. All GC samples annealed at 500°C 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°C for 10 hours. Fit model is the same as in Fig. 1.
Table 1: Coating loss, fit parameters ($\phi_c = a \cdot f^b$), refractive index $n$ at $\lambda = 1064$ nm and density $\rho$ of materials deposited in different coaters.

|        | SiO$_2$ | Ta$_2$O$_5$ |
|--------|---------|-------------|
| a ($10^{-4}$) | 1.1 ± 0.2 | 1.89 ± 0.09 |
| b ($10^{-1}$) | -0.4 ± 0.3 | 1.00 ± 0.05 |
| n | 1.474 ± 0.005 | 2.03 ± 0.02 |
| $\rho$ [g/cm$^3$] | 2.34 ± 0.01 | 7.34 ± 0.07 |

Figure 5: (Color online) Effect of annealing duration $\Delta t$ [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 $\Delta t$, avoiding coating crystallization which would increase optical loss by scattering and absorption. In Fig. 5 is shown the effect of increasing $\Delta t$ 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$_2$O$_5$; bottom row: c) structure of SiO$_2$, d) structure of Ta$_2$O$_5$.

Figure 7: (Color online) Loss of Ta$_2$O$_5$ coatings deposited by different coaters and annealed at 500°C 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 D2 has been found, suggesting that SiO2 loss increases with the 3-fold ring population [9]. This correlation holds for different kinds of SiO2, coating and bulk (Fig. 8). On the other hand, Ta2O5 loss does not change for ∆t ≥ 10h and its structure evolves only for ∆t ≤ 10h. Fig. 6 shows coating loss and structure for increasing Ta, with ∆t = 10h constant. SiO2 loss decreases and its structure evolves considerably. Surprisingly, crystallization occurs at Ta = 1000°C. For Ta2O5, coating loss is roughly constant for Ta > 500°C and its structure does not change up to Ta = 600°C, when crystallization occurs.

3. New material
TiO2:Ta2O5 could be replaced by a material with lower mechanical loss and possibly higher refractive index. Here silicon nitride (Si3N4) is proposed, which features high refractive index [12] and very low mechanical loss [13]. Usually, Si3N4 is deposited by low pressure chemical vapour deposition (LPCVD). However, LPCVD Si3N4 might suffer from hydrogen contamination and thickness uniformity issues, which are not compatible with the stringent optical specifications required for GW detectors. Instead, IBS Si3N4 can be developed in the GC for deposition on large optics. Fig. 9 shows a comparison between TiO2:Ta2O5 and IBS Si3N4, this latter being annealed at different temperatures. Si3N4 loss decreases significantly at Ta = 900°C. Thus, one could increase the annealing temperature of the entire HR stack to decrease also SiO2 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 TiO2 content in TiO2:Ta2O5. 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 SiO2. In the case of Ta2O5, the effect of different deposition parameters is erased by the first few hours of post-deposition annealing. For 400°C < Ta < 600°C and 10h < ∆t < 50h Ta2O5 loss shows null or limited evolution and its structure is frozen in a stable configuration. In the case of SiO2, the annealing parameters Ta and ∆t have a significant impact on mechanical loss and coating structure. A correlation is found between D2 peak area in the Raman spectra, associated to the three fold ring population, and mechanical loss. IBS Si3N4 is an interesting new possibility to replace TiO2:Ta2O5 because of its low mechanical loss. Furthermore, Si3N4 can be annealed at higher temperature than TiO2:Ta2O5, reducing also SiO2 coating loss angle and thus the loss of the whole HR coating.

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