Ternary Quarter Wavelength Coatings
for Gravitational Wave Detector Mirrors:
Design Optimization via Exhaustive Search

V. Pierro, V. Fiumara, F. Chiadini, V. Granata, O. Durante, J. Neilson, C. Di Giorgio, R. Fittipaldi, G. Carapella, F. Bobba, M. Principe, and I. M. Pinto

1 Dip. di Ingegneria, DING, Università del Sannio, I-82100 Benevento, Italy.
2 INFN, Sezione di Napoli Gruppo Collegato di Salerno, I-80126 Napoli, Italy.
3 Scuola di Ingegneria, Università della Basilicata, I-85100 Potenza, Italy.
4 Dip. di Ingegneria Industriale, DIIN, Università di Salerno, I-84084 Fisciano, Salerno, Italy.
5 CNR-SPIN, c/o Università di Salerno, I-84084 Fisciano, Salerno, Italy.
6 Dip. di Fisica “E.R. Casaniello”, Università di Salerno, I-84084 Fisciano, Salerno, Italy.
7 Museo Storico della Fisica e Centro Studi e Ricerche “Enrico Fermi”, I-00184 Roma, Italy.

Multimaterial optical coatings are a promising viable option to meet the challenging requirements (in terms of transmittance, absorbance and thermal noise) of next generation gravitational wave detector mirrors. In this paper we focus on ternary coatings consisting of quarter-wavelength thick layers, where a third material ($H'$) is added to the two presently in use, namely Silica ($L$) and Titania-doped Tantala ($H$), featuring higher dielectric contrast (against Silica), and lower thermal noise (compared to Titania-doped Tantala), but higher optical losses. We seek the optimal material sequences, featuring minimal thermal (Brownian) noise under prescribed transmittance and absorbance constraints, by exhaustive simulation over all possible configurations, for different values (in a meaningful range) of the optical density and extinction coefficient of the third material. In all cases studied, the optimal designs consist of a stack of ($H'$|$L$) doublets topped by a stack of ($H$|$L$) doublets, confirming previous heuristic assumptions, and the achievable coating noise power spectral density reduction factor is $\sim 0.5$. The robustness of the found optimal designs against layer thickness deposition errors and uncertainties and/or fluctuations in the optical losses of the third material is also investigated. Possible margins for further thermal noise reduction by layer thickness optimization, and strategies to implement it, are discussed.

Keywords: Dielectric Mirrors, Gravitational Wave Detectors, Multimaterial Coatings, Optimization, Thermal Noise, Thin Films

*pierro@unisannio.it*
I. INTRODUCTION

The visibility distance of the currently operating interferometric detectors of gravitational waves is set by thermal (Brownian) noise in the highly-reflective dielectric-multilayer coated mirrors of their optical cavities [1].

During the last two decades, the quest of coating materials featuring large dielectric contrast, low optical absorption (and scattering), and low mechanical losses (directly related to thermal noise, in view of the fluctuation-dissipation principle) has been quite intense.

Since the development in 1997 of Titania-doped Tantala by LMA [2], still used, together with Silica, in the mirrors of the (currently operational) ”advanced” LIGO and Virgo detectors, several potentially interesting coating materials have been investigated [see [3] for a recent review].

None of them qualifies as a straight substitute of the materials currently in use; but a few of them are better in terms of some properties (e.g., optical density, and/or mechanical losses), while unfortunately worse as regards others.

Amorphous Silicon (aSi), in particular, has received much attention, in view of its large refractive index and limited mechanical losses, down to cryogenic temperatures [4]. Its optical losses, on the other hand, are sensibly larger than those of Titania-doped Tantala, and appear to be strongly dependent on the deposition technology [5]. Amorphous Silicon has been indicated as a candidate coating material for 3rd generation (3G) cryogenic detectors using crystalline Silicon for the mirror substrate (presently made of fused Silica), and a 1550nm laser source [6].

Silicon Nitrides, (SiNx) have also been proposed as potentially interesting materials, in view of their flexible stoichiometry, that allows to tune their refractive index in a wide range of values, ability to accommodate large substrates, via plasma-enhanced chemical vapor deposition (PECVD), and pretty low mechanical losses, at ambient as well as cryo temperatures [7]. Their optical losses, though, exceed those of currently used materials [8].

In [9] and [10] it was first suggested to modify the simplest binary coating design consisting of alternating quarter-of-wavelength-(QWL)-thick layers of SiO2 and TiO2 :Ta2O5, by using a third denser but optically lossier material, in the high-index layers closest to the substrate, where the field intensity is usually low enough to make its relatively large optical losses irrelevant, to reduce the total number of layers (and hence the coating thermal noise), in view of its larger optical contrast with the low-index material. The feasibility of aSi based ternary coatings has been recently demonstrated [11].

Optical coatings using more than two lossy materials (m-ary coatings, with m > 2) have been
studied since long (see, e.g. [12] for a review). However, the design constraints/requirements of the mirrors used by interferometric detectors of gravitational waves are peculiar, especially as regards the key figure of merit represented by thermal (Brownian) noise, making further analysis necessary.

In this paper we implement exhaustive simulations aimed at identifying the structure of the optimal coating design using three different materials yielding the lowest thermal noise under prescribed (upper) bounds on power transmittance and absorbance, without any a-priori assumption, except that all layers are QWL.

The paper is organized as follows. In Section II we summarize the relevant modeling assumptions used; in Section III we introduce the exhaustive procedure used to find the optimum material sequences; in Section IV we present and discuss the simulations done, including the putative 3rd material properties used, the structure of the coating design families that comply with the prescribed transmittance and absorbance constraints, and the properties of the minimum thermal noise (optimal) ones, including robustness against layer thickness errors, and uncertainties or fluctuations in the 3rd material extinction coefficient. Possible margins for further thermal noise reduction by layer thickness optimization, and strategies to implement it, are also discussed. Conclusions follow under Section V.

II. COATING MODEL

In this section we summarize the modeling assumptions used throughout the paper, based on the transmission matrix formalism (see, e.g., [13], [14]) and the simplest thermal noise model introduced in [15].

A. Optical Modeling

Here an \( \exp(i 2\pi f_0 t) \) time-dependence is understood, \( f_0 \) being the laser light frequency and \( i \) the imaginary unit. The optical properties of a multilayer coating can be deduced from its characteristic matrix

\[
T = T_1 \cdot T_2 \cdot \ldots \cdot T_{N_T}
\]  

(1)
where $N_T$ is the total number of layers (numbered from the vacuum to the substrate as in Figure 1) and $T_m$ is the transmission matrix of the $m$-th layer. Assuming normal incidence \cite{13},

$$T_m = \begin{bmatrix} \cos (\psi_m) & (i/n^{(m)}) \sin (\psi_m) \\ n^{(m)} \sin (\psi_m) & \cos (\psi_m) \end{bmatrix}, \quad (2)$$

where

$$\psi_m = \frac{2\pi}{\lambda_0} n^{(m)} d_m, \quad (3)$$

$\lambda_0$ and $d_m$ being the light free-space wavelength and the $m$-th layer thickness, respectively, and

$$n^{(m)} = n_r^{(m)} - i\kappa^{(m)}, \quad (4)$$

$n_r^{(m)}$ being the real refractive index and $\kappa^{(m)}$ the extinction coefficient of the $m$-th layer material.

The coating is placed between two homogeneous non dissipative dielectric half-spaces with refractive indexes $n^{(0)}$ and $n_S$, respectively (see Figure 1). The bottom half-space is the substrate; the top will be assumed to be the vacuum, with $n^{(0)} = 1$.

The effective complex refractive index of the whole substrate-terminated coating is,

$$n_C = \frac{T_{21} + n_S T_{22}}{T_{11} + n_S T_{12}}, \quad (5)$$

that can be used to compute the (monochromatic plane wave, normal incidence) coating reflection coefficient $\Gamma_C$:

$$\Gamma_C = 1 - n_C \frac{1 + n_C}{1 - n_C}, \quad (6)$$

and the power-transmittance

$$\tau_C = \frac{P_m}{P^+} = 1 - |\Gamma_c|^2 \quad (7)$$

where $P_m$ is the power density flowing into the coating through the vacuum/coating interface, and $P^+$ is the power density of the incident wave,

$$P^+ = \frac{1}{2Z_0} |E_{inc}|^2 \quad (8)$$

where $E_{inc}$ is the (transverse) incident electric field at the vacuum/coating interface and $Z_0 = \sqrt{\mu_0/\epsilon_0}$ is the characteristic impedance of the vacuum.

The average power density dissipated in the coating is the difference between $P_m$ and the power density flowing into the substrate $P_{out}$. This latter can be computed as

$$P_{out} = \frac{1}{2} \Re(E^{(S)}H^{(S)*}) \quad (9)$$
where $\text{Re}(\cdot)$ gives the real part of its argument, and $E^{(S)}$ and $H^{(S)}$ are the (transverse) electric and magnetic fields at the coating/substrate interface, that are readily obtained from the fields $E^{(0)} = E_{\text{inc}}(1 + \Gamma_c)$ and $Z_0H^{(0)} = E_{\text{inc}}(1 - \Gamma_c)$ at the vacuum/coating interface using the formula

$$\begin{bmatrix} E^{(S)} \\ Z_0H^{(S)} \end{bmatrix} = T^{-1} \begin{bmatrix} E^{(0)} \\ Z_0H^{(0)} \end{bmatrix}.$$  \hfill (10)

Accordingly, the coating absorbance is

$$\alpha_C = \frac{(P_{\text{in}} - P_{\text{out}})}{P^+} = \tau_C - \tau_S$$  \hfill (11)

where

$$\tau_S = \frac{P_{\text{out}}}{P^+}$$  \hfill (12)

is the fraction of the incident power leaking into the substrate. Note that eq. (11) entails the obvious condition $\tau_C \geq \tau_S$.

B. Thermal Noise Modeling

The frequency dependent power spectral density $S^{(B)}_{\text{coat}}(f)$ of the coating thermal noise can be written

$$S^{(B)}_{\text{coat}}(f) \propto \frac{T}{w_f} \phi_c$$  \hfill (13)

where $f$ is the frequency, $T$ is the (absolute) temperature, $w$ is the (assumed Gaussian) laser-beam waist, and $\phi_c$ is the coating loss angle. Neglecting higher order terms stemming from subtler effects [16], this latter can be written [15]

$$\phi_C = \sum_{m=1}^{N_T} \eta_m d_m$$  \hfill (14)

where

$$\eta_m = \frac{1}{\sqrt{\pi w}} \phi_m \left( \frac{Y_m}{Y_S} + \frac{Y_S}{Y_m} \right)$$  \hfill (15)

is the specific loss angle (loss angle per unit thickness) of the material making the $m$–th layer, $\phi_m$ and $Y_m$ being its mechanical loss angle and Young’s modulus, respectively, and $Y_S$ the Young modulus of the substrate.

According to eq. (13), lowering the temperature $T$ would reduce thermal noise [32]. However, in many coating materials, including those currently in use (Silica and Titania-doped Tantala), mechanical-losses peak [18] [19] in the range of cryo-temperatures of interest for next generation detectors like ET [20] and LIGO-CE [21], pioneered by KAGRA [22].
III. EXHAUSTIVE SCRUTINY OF QWL TERNARY COATINGS

The exhaustive scrutiny consists in evaluating the performance (in terms of power transmittance, power absorbance and coating loss angle) of all (admissible) ternary coatings consisting of QWL layers made of three possible materials henceforth denoted as $L$, $H$ and $H'$, that comply with given transmittance and absorbance constraints,

$$\tau_C \leq \tau_{ref}, \quad \alpha_C \leq \alpha_{ref}$$  \hspace{1cm} (16)

For coatings consisting of QWL layers, the matrices $T_m$, $m = 1, 2, \ldots, N_T$ in (2) take the simple form

$$T_m = \begin{bmatrix}
\sinh \left( \frac{\pi \kappa^{(m)}}{2 n_r^{(m)}} \right) & \frac{1}{n^{(m)}} \cosh \left( \frac{\pi \kappa^{(m)}}{2 n_r^{(m)}} \right) \\
n^{(m)} \cosh \left( \frac{\pi \kappa^{(m)}}{2 n_r^{(m)}} \right) & \sinh \left( \frac{\pi \kappa^{(m)}}{2 n_r^{(m)}} \right)
\end{bmatrix}.$$  \hspace{1cm} (17)

Note that expansion of the hyperbolic functions to 1st order in $\kappa^{(m)}$ (assumed $\ll 1$ for all materials involved) may lead to inaccuracies in the computed absorbances that could be non-negligible comparable to the enforced absorbance bounds.

For ternary coatings $n^{(m)}$ can only take values in $\{n_L, n_H, n_H'\}$, and the corresponding single-layer matrices will be denoted as $T_L$, $T_H$ and $T_{H'}$, respectively.

Among all possible material sequences, those for which $n^{(m)} = n^{(m-1)}$ for some $m$ should be obviously discarded [33]. Accordingly, we are left with a total of $N_C = 3 \cdot 2^{N_T-1}$ distinct acceptable ternary coatings consisting of $N_T$ QWL layers [34].

Knowledge of the matrix [1] yields the coating transmittance and absorbance, as shown in Sect. [11]

In order to compute the coating thermal noise it is expedient to let:

$$\gamma_H = \frac{\text{Re}[n_L]}{\text{Re}[n_H]} \frac{\eta_H}{\eta_L} = \frac{\text{Re}[n_L]}{\text{Re}[n_H]} \frac{\phi_H}{\phi_L} \left( \frac{Y_H + Y_s}{Y_H} \right) \left( \frac{Y_L + Y_s}{Y_L} \right)^{-1}$$  \hspace{1cm} (18)

$$\gamma_{H'} = \frac{\text{Re}[n_L]}{\text{Re}[n_{H'}]} \frac{\eta_{H'}}{\eta_L} = \frac{\text{Re}[n_L]}{\text{Re}[n_{H'}]} \frac{\phi_{H'}}{\phi_L} \left( \frac{Y_H + Y_s}{Y_{H'}} \right) \left( \frac{Y_L + Y_s}{Y_L} \right)^{-1}$$  \hspace{1cm} (19)

where $\phi_L$, $\phi_H$ and $\phi_{H'}$ are the material mechanical loss angles, whereby, using (14) and (15)

$$\phi_C = \frac{\eta_L \lambda_0}{4 \text{Re}(n_L)} (N_L + \gamma_H N_H + \gamma_{H'} N_{H'})$$  \hspace{1cm} (20)
$N_L$, $N_H$ and $N_{H'}$ being the number of layers made of the $L$, $H$, and $H'$ materials, respectively.

The above is a typical constrained optimization problem \cite{23}, and has combinatorial complexity. In order to keep the computational burden and computing times within acceptable limits, we use the backtracking strategy \cite{24} to reduce the number of matrix multiplications.

\section{NUMERICAL EXPERIMENTS}

In this Section we apply the above mentioned exhaustive scrutinizing procedure to ternary QWL coatings laid on a fused-Silica substrate (assumed of infinite thickness), using SiO$_2$ and TiO$_2$ :: Ta$_2$O$_5$ for the low ($L$) and high ($H$) index materials, respectively. For illustrative purposes, we shall consider two putative candidates for the 3rd (high-index) material ($H'$). These will be referred to as Material-A and Material-B.

Material-A has the same mechanical losses as TiO$_2$ :: Ta$_2$O$_5$, a fairly higher refractive index, but larger optical losses; Material-B has the same refractive index as TiO$_2$ :: Ta$_2$O$_5$, fairly lower mechanical losses, but larger optical losses. Materials A and B are loosely resemblant of amorphous Silicon and Silicon Nitride, respectively.

For both materials we consider values of the extinction coefficient ranging from $10^{-6}$ to $10^{-4}$ in decadic steps.

The numerical values of the material properties used in our simulations were taken from \cite{9}, and are collected in Table-I.

We used the following bounds in the transmittance and absorbance constraints \cite{16}:

$$\tau_{\text{ref}} = 6 \text{ ppm}, \quad \alpha_{\text{ref}} = 1 \text{ ppm},$$

and scaled the loss angle of the various admissible solutions to that of a reference LIGO/Virgo-like design, consisting of $N_T = 36$ alternating Titania-doped-Tantala/Silica layers, for which

$$\phi_C = \phi_{C}^{(\text{ref})} = 18 \frac{\eta L \lambda_0}{4 Re(n_L)} (1 + \gamma_H).$$

\subsection{Ternary QWL Coatings Using Material-A}

The set of all admissible ternary QWL coatings compliant with the prescribed transmittance and absorbance constraints \cite{21} and using Material-A for $H'$ is collected in Table II for three possible values of the extinction coefficient ($\kappa_{H'} = 10^{-6}, 10^{-5}, 10^{-4}$).
They are conveniently grouped into subsets featuring the same number of \([H|L]\), \([H'|L]\) and \([H'|H]\) doublets, and hence the same coating loss angle, in order of increasing transmittance and/or absorbance. For all considered \(\kappa_{H'}\) values, the optimal design featuring the lowest coating loss angle consists of a stack of \(N_{H'}\) doublets \([H'|L]\) grown on top of the substrate, topped by another stack of \(N_H\) doublets \([H|L]\), where the optimal values of \(N_{H'}\) and \(N_H\) depend on the extinction coefficient of the \(H'\) material.

There are no \([H'|H]\) (nor \([H|H']\)) doublets in the optimal designs.

These findings confirm the heuristic assumption first made in [9], [10] about the structure of ternary QWL coatings yielding minimal noise under prescribed transmittance and absorbance constraints.

**B. Ternary QWL Coatings Using Material-B**

In this case, materials \(H\) and \(H'\) are iso-refractive, hence the total number of high-low index doublets needed to satisfy the prescribed transmittance constraint remains fixed, irrespective of whether the high index layers consist of the \(H\) or \(H'\) material, and is the same as for the reference \(\text{TiO}_2:\text{Ta}_2\text{O}_5/\text{SiO}_2\) binary coating. Hence,

\[
N_H + N_{H'} = N_L = N_L^{(ref)}
\]  

(23)

Note also that in this case \([H'|H]\) (or \([H|H']\)) doublets are forbidden, being optically homogenous and half-wavelength thick.

The set of all admissible ternary QWL coating compliant with the prescribed transmittance and absorbance constraints [21] can be conveniently visualized as in Figure 2, that refers to the case \(\kappa_{H'} = 10^{-5}\). It is seen that all sub-optimal, constraint-compliant admissible designs can be divided into distinct families, represented by the aligned markers in Figure 2 (bottom), each family featuring a number \((N_H)\) of \([H|L]\) doublets (denoted as \(#HL\) in figure 2), and a fixed number \((N_{H'} = N_L - N_H)\) of \([H'|L]\) doublets (denoted as \(#H'L\) in figure 2), and (hence) the same loss-angle, but different transmittances and absorbances, as seen from from Figure 2 (top).

For each family, the number of distinct admissible designs, in square brackets in Figure 2 (bottom) is (slightly) less than the binomial coefficient

\[
\binom{N_H}{N_L - N_H}
\]

(24)

due to the (relatively few) designs that do not fulfill the transmittance/absorbance constraints.
Similar to the previous case, the optimal design featuring the lowest thermal noise under the prescribed transmittance and absorbance constraints, consists of a stack of \([H'|L]\) doublets grown on top of the substrate, topped by another stack of \([H|L]\) doublets, again confirming the ansatz in [9], [10].

The found optimal designs using materials A and B for \(H'\), with \((\kappa_{H'} = 10^{-6}, 10^{-5}, 10^{-4})\) are displayed in Figure 3.

C. Robustness

The optimal ternary QWL coatings are nicely robust against uncertainties in the value of the extinction coefficient \(\kappa_{H'}\), as well as against unavoidable inaccuracies in the layers’ thicknesses, due to technological limitations of the deposition process.

As an illustration, Figure 4 shows the distributions of coating transmittances and absorbances in \(10^5\) realizations of the optimal ternary QWL coating using Material-A.

The left-column panels refer to the case i) where the extinction coefficient \(\kappa_A\) is the same for all \(H'\) layers, and is random uniformly distributed in \((0.5\bar{\kappa}_A, 1.5\bar{\kappa}_A)\), with \(\bar{\kappa}_A = 10^{-4}\); the right column panels refer to the case ii) where the extinction coefficients of the \(H'\) layers are independent random variables, identically distributed as in case i).

Similarly, Figure 5 shows the distributions of coating transmittance and absorbance in a sample of \(10^5\) realizations of the optimal ternary QWL coating using Material-B, assuming \(\kappa_B\) to be random uniformly distributed in \((0.5\bar{\kappa}_B, 1.5\bar{\kappa}_B)\), with \(\bar{\kappa}_B = 10^{-5}\).

Not unexpectedly, uncertainties in the extinction coefficient stemming from fluctuations in the deposition process, rather than systematic uncertainty in the nominal value, have a lesser effect, due to possible fluctuations compensation, resulting into narrower distributions of the coating transmittance and absorbance.

Figure 6 shows the distributions of coating transmittance, absorbance and loss angle (normalized to the value of the reference binary coating) in a sample of \(10^5\) realizations of i) the optimal ternary QWL coating using Material-A for \(H'\), with \(\kappa_A = 10^{-4}\) (left panels), and ii) the optimal ternary QWL coating using Material-B for \(H'\), with \(\kappa_B = 10^{-5}\) (right panels), assuming the thicknesses of all layers to be independent random variables identically distributed uniformly around the nominal QWL thickness, in a symmetric interval of total width \(2\text{nm}\).

We may conclude that the optimal ternary QWL designs are fairly robust against uncertainties in the extinction coefficient of the \(H'\) material, and deposition-related thickness errors.
D. Transmittance Spectra

We computed the transmittance spectra of the above optimal ternary QWL coatings. These are shown in Figure 7.

The spectra were computed neglecting chromatic dispersion, except in a neighbourhood of the operating wavelength ($\lambda_0 = 1064$ nm), shown in the insets, where a linear approximation was used,

$$n_r(\lambda) = n_r(\lambda_0) + \frac{dn_r}{d\lambda} \bigg|_{\lambda_0} (\lambda - \lambda_0).$$

(25)

The pertinent values of the $\frac{dn_r}{d\lambda} \bigg|_{\lambda_0}$ were taken from [25], and are: $-1.2 \cdot 10^{-5}$ nm$^{-1}$ for Silica, $-4.9 \cdot 10^{-5}$ nm$^{-1}$ for Titania-doped Tantala, $-3.8 \cdot 10^{-5}$ nm$^{-1}$ for material-A and $-4.26 \cdot 10^{-4}$ for material-B.

Within the limits of this model, no ripple is observed in the high-reflectance band. The shape of the transmission spectrum for the optimal coating using material-A departs more markedly from the reference spectrum, compared to that of the optimal coating using material-B. The observed asymmetry of the lobes stems from the fact that the coating is piecewise homogeneous, consisting of two cascaded homogeneous QWL stacks.

E. Thickness Optimization

Thermal noise in binary coatings can be effectively reduced, compared to the reference QWL-layer design, by suitably reducing the total thickness of the optically noisier material(s), while increasing the total thickness of the other material(s), and the total number of doublets, so as to keep the coating transmittance unchanged [14].

Remarkably, the thickness-optimized binary coatings turn out to consist of almost identical stacked $[H|L]$-doublets whose thickness is one half of the working wavelength (Bragg condition), the exception being represented by a fewest layers near the coating top and bottom [26], [27], [28], [29].

Implementing thickness optimization for ternary coatings is computationally demanding. In a full-blind exhaustive approach, each and any $L$ layer should be allowed to take any thickness value in the range $(\lambda/4, \lambda/2)$, and each and any $H$ and $H'$ layer should be allowed to take any thickness in $(0, \lambda/4)$, $\lambda$ being the local wavelength. Even after suitable discretization of the above search intervals, the computational burden of an exhaustive search would become prohibitive for any meaningful value of $N_T$. 
A reasonable heuristic approach to thickness optimization of ternary QWL coatings may thus consist in optimizing the two binary QWL stacks that form the top and bottom part of the optimal QWL ternary coatings, assuming each of them to consist of identical non-QWL Bragg doublets [30].

Here, to illustrate the possible margins of further loss angle reduction obtainable from thickness optimization, we content ourselves to compute the (normalized) coating loss angle, absorbance and power transmittance of the coatings obtained after modifying the optimal ternary QWL coatings found in the previous Sections and listed in Figure 3 by letting $n_H' d_H' = \frac{1}{6}$, and $n_L d_L = \frac{1}{3}$ (26) so as to preserve the Bragg character of the doublets, and adding a few $[H|L]$ and/or $[H'L]$ layers as needed to maintain compliance with the transmittance and absorbance constraints.

The transmittance, absorbance and coating loss angle (normalized to the reference value) of the resulting thickness-tweaked coatings are collected under Table-III.

V. CONCLUSIONS

In this paper we addressed the problem of designing a ternary optical coating consisting of QWL layers to achieve the minimum thermal (Brownian) noise under prescribed optical transmittance and absorbance constraints.

We considered ternary coatings where two materials are those presently in use in the advanced LIGO and Virgo detectors, namely SiO$_2$ and TiO$_2$ :: Ta$_2$O$_5$, featuring the best tradeoff between optical contrast, optical losses and thermal noise so far; and the third material is either similar to $\alpha$Si, featuring the same mechanical losses as TiO$_2$ :: Ta$_2$O$_5$, a higher refractive index, but larger optical losses; or to SiN$_x$, featuring the same refractive index as TiO$_2$ :: Ta$_2$O$_5$, fairly lower mechanical losses, but larger optical losses.

In order to take into account the present uncertainties (and possible margins of improvement [31]) for the optical losses of these materials, we considered different values of their extinction coefficient ranging from $10^{-6}$ to $10^{-4}$.

We performed an exhaustive search over all possible (and admissible, in the sense discussed in Sect. IV) configurations consisting of QWL layers, using backtracking for numerical efficiency, seeking for the design yielding minimum thermal (Brownian) noise under prescribed upper bounds for transmittance and absorbance.
The main results of this study can be summarized as follows.

All optimal designs, featuring the lowest thermal noise under the prescribed transmittance and absorbance constraints, in the considered range of material parameters, consist of a stack of \([H'|L]\) doublets grown on top of the substrate, topped by another stack of \([H|L]\) doublets, confirming the ansatz in \([9], [10]\).

All the optimal solutions outperform significantly the reference binary solution consisting of alternating QWL layers made of Silica and Titania-doped Tantala, in terms of Brownian thermal noise. The coating loss angle reduction increases as the the extinction coefficient of the added (third) material is decreased.

The found optimal solution are nicely robust against deposition inaccuracies in the individual layer thicknesses, and systematic uncertainties and/or fluctuations from layer-to-layer, of the extinction coefficient. The transmittance spectra have been computed, and satisfy the design constraints in the useful band.

Finally, we have shown that a further improvement in performance can be achieved by using thinner layers of the noisier materials, in each Bragg doublet.

Exhaustive blind thickness optimization of ternary coatings appears to be computationally unaffordable. The problem will be accordingly studied in a future paper, following a heuristic approach.

ACKNOWLEDGEMENTS

This work has been supported in part by INFN (Istituto Nazionale di Fisica Nucleare) through the projects Virgo and ET-Italy, and by the European Gravitational Observatory (EGO). The authors gratefully acknowledge useful discussions with, and suggestions from members of the Virgo Coating R & D Group (VCR & D) and the LIGO Optics Working Group (OWG).
\[ n^{(0)} = 1 \] (vacuum)

\[ n^{(1)} \quad n^{(2)} \quad \ldots \]

\[ d_1 \quad d_2 \quad \ldots \quad d_{N_T} \]

\[ 1 \quad 2 \quad \ldots \quad N_T \]

FIG. 1: Sketch of a multilayer coating.
| Property | SiO$_2$ | TiO$_2$:Ta$_2$O$_5$ | MA      | MB      |
|----------|---------|---------------------|---------|---------|
| $n_r$    | 1.45    | 2.1                 | 3.0     | 2.1     |
| $\kappa$| $10^{-11}$ | $2 \times 10^{-8}$ | $10^{-6}$ to $10^{-4}$ | $10^{-6}$ to $10^{-4}$ |
| $Y$      | 72 GPa  | 140 GPa             | 100 GPa | 100 GPa |
| $\phi$  | $5.0 \times 10^{-5}$ | $3.76 \times 10^{-4}$ | $3.76 \times 10^{-4}$ | $1 \times 10^{-4}$ |

TABLE I: Numerical values of coating material parameters used in simulations. All symbols have the usual meaning.
\[ \kappa_{H'} = 10^{-6} \]

| Structure | \( \tau_C \) [ppm] | \( \alpha_C \) [ppm] | \( \phi_C/\phi_C^{(ref)} \) |
|-----------|-----------------|-----------------|-----------------|
| (#HL, #H'L, #H'H) = (0,10,0) | 2.25 | 0.911 | 0.377 |
| (#HL, #H'L, #H'H) = (1,9,0) | 3.19 | 0.463 | 0.395 |
| (#HL, #H'L, #H'H) = (2,8,0) | 5.81 | 0.249 | 0.412 |
| (#HL, #H'L, #H'H) = (3,7,0) | 5.97 | 0.413 | 0.412 |
| (#HL, #H'L, #H'H) = (4,7,0) | 5.934 | 0.522 | 0.486 |
| (#HL, #H'L, #H'H) = (5,6,0) | 5.960 | 0.695 | 0.656 |
| (#HL, #H'L, #H'H) = (6,5,0) | 5.960 | 0.695 | 0.656 |
| (#HL, #H'L, #H'H) = (7,5,0) | 5.960 | 0.695 | 0.656 |

\[ \kappa_{H'} = 10^{-5} \]

| Structure | \( \tau_C \) [ppm] | \( \alpha_C \) [ppm] | \( \phi_C/\phi_C^{(ref)} \) |
|-----------|-----------------|-----------------|-----------------|
| (#HL, #H'L, #H'H) = (0,9,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (1,8,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (2,7,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (3,6,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (4,6,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (5,5,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (6,5,1) | 3.712 | 0.911 | 0.418 |
| (#HL, #H'L, #H'H) = (7,5,1) | 3.712 | 0.911 | 0.418 |

\[ \kappa_{H'} = 10^{-4} \]

| Structure | \( \tau_C \) [ppm] | \( \alpha_C \) [ppm] | \( \phi_C/\phi_C^{(ref)} \) |
|-----------|-----------------|-----------------|-----------------|
| (#HL, #H'L, #H'H) = (6,5,0) | 5.420 | 0.297 | 0.633 |
| (#HL, #H'L, #H'H) = (7,5,0) | 5.687 | 0.564 | 0.633 |
| (#HL, #H'L, #H'H) = (8,4,0) | 5.687 | 0.564 | 0.633 |

**TABLE II:** The set of all admissible ternary QWL coatings subject to (16) using Silica (L), Titania-doped Tantala (H) and material A for (H'), with \( \kappa_{H'} = 10^{-6}, 10^{-5}, 10^{-4} \). See Section III for details.
FIG. 2: The set of all admissible ternary QWL coatings subject to (16) using Silica (L), Titania-doped Tantala (H) and material B for (H′), with $\kappa_{H'} = 10^{-5}$. See Section III for details.
\[ \kappa = 10^{-6}, N_T = 20 \]
\[ \kappa = 10^{-5}, N_T = 22 \]
\[ \kappa = 10^{-4}, N_T = 26 \]

**Optimal ternary QWL coatings using Material-A**

| \( \kappa \)  | \( \tau_C \) [ppm] | \( \alpha_C \) [ppm] | \( \phi_C/\phi_f \) |
|----------------|---------------------|---------------------|---------------------|
| 10^{-6}       | 2.246               | 0.911               | 0.377               |
| 10^{-5}       | 5.934               | 0.522               | 0.486               |
| 10^{-4}       | 3.074               | 0.564               | 0.615               |

**Legend:** L= ; H= ; H'= 

\[ \kappa = 10^{-6}, N_T = 36 \]
\[ \kappa = 10^{-5}, N_T = 36 \]
\[ \kappa = 10^{-4}, N_T = 36 \]

**Optimal ternary QWL coatings using Material-B**

| \( \kappa \)  | \( \tau_C \) [ppm] | \( \alpha_C \) [ppm] | \( \phi_C/\phi_f \) |
|----------------|---------------------|---------------------|---------------------|
| 10^{-6}       | 5.128               | 0.661               | 0.401               |
| 10^{-5}       | 5.191               | 0.724               | 0.514               |
| 10^{-4}       | 5.247               | 0.781               | 0.626               |

FIG. 3: The found optimal (minimum Brownian noise) QWL ternary coating designs subject to \([16]\), using Silica (L), Titania-doped Tantala (H) and materials A and B (H'), assuming different values for \( \kappa_{H'} \) (substrate on the right).
FIG. 4: Distributions of coating transmittance and absorbance in a sample of $10^5$ realizations of the optimal ternary QWL coating using Material-A, assuming $\kappa_A$ to be random uniform in $(0.5\bar{\kappa}_A, 1.5\bar{\kappa}_A)$, with $\bar{\kappa}_A = 10^{-4}$. The left-column panels refer to the case where the extinction coefficient is the same for all $H'$ layers; the right column panels refer to the case ii) where the extinction coefficients of the $H'$ layers are independent identically distributed random variables.
Optimal ternary coating using Material-B

FIG. 5: Distributions of coating transmittance and absorbance in a sample of $10^5$ realizations of the optimal ternary QWL coating using Material-B, assuming $\kappa_B$ to be random uniform in $(0.5\bar{\kappa}_B, 1.5\bar{\kappa}_B)$, with $\bar{\kappa}_B = 10^{-5}$. The left-column panels refer to the case where the extinction coefficient is the same for all $H'$ layers; the right column panels refer to the case ii) where the the extinction coefficients of the $H'$ layers are independent identically distributed random variables.
FIG. 6: Distributions of coating transmittance, absorbance and loss angle (normalized to the value of the reference binary coating) in a sample of $10^5$ realizations, assuming the thicknesses of all layers to be independent random variables identically distributed uniformly around the nominal QWL thickness, in a symmetric interval of total width $2nm$. Left panels: optimal ternary QWL coating using Material-A for $H'$, with $\kappa_A = 10^{-4}$. Right panels: optimal ternary QWL coating using Material-B for $H'$, with $\kappa_B = 10^{-5}$. 
FIG. 7: Transmittance spectra of optimal designs (solid curve) using for $H'$ material-A for, with $\kappa_{H'} = 10^{-4}$, and material-B with $\kappa_{H'} = 10^{-5}$. Close-ups shown in insets. The dashed curve indicates the performance of the reference QWL design.
TABLE III: Thickness-tweaked optimal ternary QWL designs.
[1] D.V. Martynov et al., "Sensitivity of the Advanced LIGO Detectors at the Beginning of Gravitational Wave Astronomy," Phys. Rev. D93 (2016) 112004.

[2] G.M. Harry et al., "Titania-Doped Tantala/Silica Coatings for Gravitational Wave Detectors," Class. Quantum Grav. 24 (2007) 405.

[3] G. Vajente, "Review of Amorphous Coatings," LIGO Document G1900400 (2019).

[4] J. Steinlechner et al, "Silicon-Based Optical Mirror Coatings for Ultrahigh Precision Metrology and Sensing," Phys. Rev. Lett 120 (2018) 263602.

[5] L. Terkovsky et al, "Influence of Deposition Parameters on the Optical Absorption of Amorphous Silicon Thin Films," Phys. Rev. Research 2 (2020) 033308.

[6] R. Schnabel et al., "Building Blocks for Future Detectors: Silicon Testmasses and 1550 nm Laser Light," J. Phys.: Conf. Ser. 228 (2010) 012029.

[7] H.-W. Pan et al., "Silicon Nitride and Silica Quarter-Wave Stacks for Low-Thermal-Noise Mirror Coatings," Phys. Rev. D98 (2018) 102001.

[8] J. Steinlechner et al., "Optical absorption of silicon nitride membranes at 1064 nm and at 1550 nm," Phys. Rev. D96 (2017) 022007.

[9] W. Yam, S. Gras and M. Evans "Multimaterial Coatings with Reduced Thermal Noise," Phys. Rev. D91 (2015) 042002.

[10] J. Steinlechner et al. "Thermal Noise Reduction and Absorption Optimization via Multimaterial Coatings," Phys. Rev. D91 (2015) 042001.

[11] S.C. Tait et al., "Demonstration of the Multimaterial Coating Concept to Reduce Thermal Noise in Gravitational-Wave Detectors," Phys. Rev. Lett. 125 (2020) 011102.

[12] J.I. Larruquert et al., "Constructing Multilayers with Absorbing Materials," Chinese Opt. Lett. Suppl. 8 (2010) 159, and references therein.

[13] Orfanidis S J Electromagnetic Waves and Antennas (web book, [https://www.ece.rutgers.edu/~orfanidi/ewa/](https://www.ece.rutgers.edu/~orfanidi/ewa/))

[14] I.M. Pinto, M. Principe and R. DeSalvo, "Reflectivity and Thickness Optimization," Ch. 10 in Optical Coatings and Thermal Noise in Precision Measurements, G.Harry, T. R. Bodiya and R. DeSalvo, Eds., Cambrudge Univ. Press (2012).

[15] G.M. Harry et al., "Thermal Noise from Optical Coatings in Gravitational Wave Detectors," Appl. Opt. 45 (2006) 1569.

[16] T. Hong et al., "Gravitational Wave Detectors," Phys. Rev. D87 (2013) 82001.

[17] M. Principe, "Reflective Coating Optimization for Interferometric Detectors of Gravitational Waves ," Optics Expr. 23 (2015) 10938.

[18] I. Martin et al., "Measurements of a Low-Temperature Mechanical Dissipation Peak in a Single Layer of Ta2O5 Doped with TiO2," Class. Quantum Grav., 25 (2008) 055005
[19] I. Martin et al., "Low Temperature Mechanical Dissipation of an Ion-Beam Sputtered Silica Film," Class. Quantum Grav. 31 (2014) 035019.
[20] http://www.et-gw.eu/index.php
[21] https://cosmicexplorer.org/
[22] T. Akutsu and the KAGRA Collaboration, "KAGRA: 2.5 Generation Interferometric Gravitational Wave Detector," Nature Astron., 3 (2019) 35.
[23] K. Ghedira and B. Dubuisson Constraint Satisfaction Problems (John Wiley & Sons 2013).
[24] D.E. Knuth, The Art of Computer Programming, Vol. 4 (5B) (Addison-Wesley, 1968).
[25] https://refractiveindex.info
[26] J. Agresti et al., "Optimized Multilayer Dielectric Mirror Coatings for Gravitational Wave Interferometers," in Proc. SPIE - 6286 Advances in Thin-Film Coatings for Optical Applications III, M.J. Ellison, Ed., 628608, (2006).
[27] A.E. Villar et al., "Measurement of Thermal Noise in Multilayer Coatings with Optimized Layer Thickness," Phys. Rev. D81 (2010) 122001.
[28] N. M. Kondratiev, A. G. Gurkovsky, and M. L. Gorodetsky, "Thermal Noise and Coating Optimization in Multilayer Dielectric Mirrors," Phys. Rev. D84 (2011) 022001.
[29] V. Pierro et al., "On the Performance Limits of Coatings for Gravitational Wave Detectors Made of Alternating Layers of Two Materials," Optical Mater., 96 (2019) 109269.
[30] I.M. Pinto, "Stacked-Triplet Ternary HR Coatings : Another Multimaterial Design Option," LIGO Document G2000218 (2020).
[31] R. Birney et al., "Amorphous Silicon with Extremely Low Absorption: Beating Thermal Noise in Gravitational Astronomy," Phys. Rev. Lett. 121 (2018) 191101.
[32] Other options, that also appear promising, though on different timescales, including wide beams, crystalline materials, nanolayered composites, coating-less mirrors and diffractive optics [17] fall beyond the scope of the present paper.
[33] Addition of a layer with $n^{(m)} = n^{(m-1)}$ would only increase coating thermal noise, and absorbance, while having no effect on coating transmittance, the transmission matrix of the resulting half-wavelength thick homogeneous layer being the identity matrix, as seen from eq. [2].
[34] This follows immediately by noting that the first layer can be chosen in three possible ways, while each of the $N_T - 1$ subsequent ones can be chosen only in two possible ways, in view of the condition $n^{(m)} \neq n^{(m-1)}$. 