# Silicon nitride and silica quarter-wave stacks for low-thermal-noise mirror coatings

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This study investigates a multilayer high reflector with new coating materials for next-generation laser interferometer gravitational wave detectors operated at cryogenic temperatures. We use the plasma-enhanced chemical vapor deposition method to deposit amorphous silicon nitride and silica quarter-wave high-reflector stacks and studied the properties pertinent to the coating thermal noise. Room- and cryogenic-temperature mechanical loss angles of the silicon nitride and silica quarter-wave bilayers are measured using the cantilever ring-down method. We show, for the first time, that the bulk and shear loss angles of the coatings can be obtained from the cantilever ring-down measurement, and we use the bulk and shear losses to calculate the coating thermal noise of silicon nitride and silica high-reflector coatings. The mechanical loss angle of the silicon nitride and silica bilayer is dispersive with a linear weakly positive frequency dependence, and, hence, the coating thermal noise of the high reflectors show a weakly positive frequency dependence in addition to the normal $1/\sqrt{f}$ dependence. The coating thermal noise of the silicon nitride and silica high-reflector stack is compared to the lower limit of the coating thermal noise of the end test mirrors of ET-LF, KAGRA, LIGO Voyager, and the directly measured coating thermal noise of the current coatings of Advanced LIGO. The optical absorption of the silicon nitride and silica high reflector at 1550 nm is 45.9 ppm. Using a multimaterial system composed of seven pairs of ion-beam-sputter deposited $Ti:Ta_2O_5$ and silica and nine pairs of silicon nitride and silica on a silicon substrate, the optical absorption can be reduced to 2 ppm, which meets the specification of LIGO Voyager.

## I. INTRODUCTION

After the first detection of gravitational waves from a binary black hole merger [1] by laser interferometer gravitational wave detectors and the recent observation of the complete evolution of a binary neutron star merger [2], gravitational-wave astronomy is moving forward at a fast pace. Next-generation laser interferometer gravitational wave detectors operated at cryogenic temperatures for higher sensitivity have been planned, i.e., the Einstein-telescope low frequency (ET-LF) at 10 K [3] and LIGO Voyager at 120 K [4], or are already in construction, i.e., KAGRA at 20 K [5]. Reducing the cryogenic Brownian thermal noise of the mirror coating around the most sensitive frequency range ($\approx$100 Hz) is becoming particularly critical with the advances in the reduction of quantum noise using squeezed-light technologies [6,7].

A high-reflector (HR) dielectric coating is composed of pairs of high- and low-refractive-index films, each of one quarter-wave (QW) optical thickness, deposited alternately on a substrate. We previously reported [8] the use of silicon nitride films deposited via the plasma-enhanced chemical vapor deposition (PECVD) method for mirror coatings. The physical properties of the silicon nitride films vary with the composition of the films. We found that the properties of $SiN_{0.40}H_{0.79}$, such as its high refractive index, low optical absorption, and low mechanical loss, and in particular, the absence of a cryogenic mechanical loss peak, render it most suitable for mirror coatings. According to the criteria set for the thin-film materials for application as the mirror coatings of a laser interferometer gravitational wave detector [8,9], in this work, we pair the $SiN_{0.40}H_{0.79}$ film with a silica film, both deposited via PECVD and each of one QW optical thickness, to form the basic QW high- and low-refractive-index bilayer building unit of the HR coatings. Cryogenic- and room-temperature ($\approx$290 K) mechanical losses were measured and the Brownian coating thermal noise (CTN)

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was calculated, analyzed, and compared with the specifications of ET-LF, LIGO Voyager, KAGRA, and aLIGO. Hereafter, "silicon nitride" specifically refers to $SiN_{0.40}H_{0.79}$.

## II. DEPOSITION AND BASIC PROPERTIES OF MATERIALS

Both silicon nitride and silica films were deposited by the PECVD method. The advantages of using PECVD over the conventional ion beam sputter (IBS) method include the ease of achieving large-area coating uniformity [8]. Precursors with predetermined flow rates were fed into the reaction chamber together with the carrier gas to form plasma with free radicals. Chemical reactions occurred near the substrate held at an elevated temperature, and the products condensed on the substrate to form a film. The PECVD processes for depositing silicon nitride and silica were described in the previous reports [8,10] and the process parameters are summarized in Table I.

The refractive index $n$, extinction coefficient $\kappa$, stress, Young's modulus $Y$, Poisson's ratio $\sigma$, and density $\rho$ of the films were also presented in the previous reports [8,10–13], and they are summarized in Table II. The stresses of the silicon nitride and silica films are close in magnitude but opposite in sign, indicating that the multilayer stack is less susceptible to stress incompatibility and is hence mechanically stable. The refractive index difference between the two materials is larger than that of the conventional $Ti:Ta_2O_5$ and silica QW stack as shown in Tables II and VIII, indicating that fewer QW pairs are required to achieve the same level of transmittance and it is in favor of achieving lower CTN.

Multilayer QW stacks of silicon nitride and silica were deposited in the same PECVD chamber by switching the process parameters alternately between the conditions given in Table I. Deposition rates for each material were predetermined—$3.1 \pm 0.2$ Å/s for silicon nitride and $10.7 \pm 0.2$ Å/s for silica—and the thicknesses of the films

were controlled using the deposition time. Thicknesses of the films were measured by using a transmission electron microscope and they were found to be within $\pm 2.5\%$ and $\pm 1.0\%$ of the designed values for silicon nitride and silica, respectively. The films were deposited on (001) silicon cantilevers. Cantilever samples with 1-,2-,3-,4-, and 8-pairs of silicon nitride and silica QW bilayers were deposited for the measurements of mechanical loss and the following analysis.

## III. THEORY

### A. Coating thermal noise

The power spectral density of the Brownian coating thermal noise $S_x(f)$ was derived [13] from the fluctuation-dissipation theorem [14] as follows:

$$S_x(f) = \frac{4K_B T}{\pi f} \frac{U}{F_0^2} \phi(f), \tag{1}$$

where $K_B$ is the Boltzman constant, $T$ is the temperature, $U$ is the maximum stored energy in the coating when a cyclic driving force with frequency $f$ and amplitude $F_0$ is applied, and $\phi(f)$ is the mechanical loss angle of the coating.

The maximum stored energy $U$ and the average energy dissipation per cycle $2\pi\phi U$ are composed of independent components from the bulk and shear motions as demonstrated by Hong *et al.* [15]. $\phi(f)$ and $S_x(f)$ were given as

$$\phi(f) = \frac{U_B}{U} \phi_B(f) + \frac{U_S}{U} \phi_S(f), \tag{2}$$

$$S_x(f) = \frac{4K_B T}{\pi f} \left[ \frac{U_B}{F_0^2} \phi_B(f) + \frac{U_S}{F_0^2} \phi_S(f) \right], \tag{3}$$

where $U_B$, $U_S$ and $\phi_B(f)$, $\phi_S(f)$ are the maximum stored energies and the loss angles of the bulk and shear motion, respectively.

TABLE I. PECVD process parameters: precursors, flow rate, carrier gas, substrate temperature, and total pressure for depositing silicon nitride and silica films. The RF power was 20 W at 13.56 MHz.

| Material | Precursor (sccm) | Carrier gas (sccm) | Temp. (°C) | Press. (torr) |
|---|---|---|---|---|
| $SiN_{0.40}H_{0.79}$ | $SiH_4/NH_3$ (45/15) | $N_2$ (980) | 300 | 1 |
| $SiO_2$ | $SiH_4/N_2O$ (8.5/710) | $N_2$ (161.5) | 300 | 1 |

TABLE II. Basic material properties of silicon nitride and silica films deposited via PECVD.

| Material | $n$ (1064 nm) | $n$ (1550 nm) | $\kappa$[a] (1550 nm, $10^{-5}$) | Stress (MPa) | $Y$ (GPa) | $\sigma$ | $\rho$ (g/cm$^3$) |
|---|---|---|---|---|---|---|---|
| $SiN_{0.40}H_{0.79}$ [8] | $2.30 \pm 0.01$ | $2.28 \pm 0.01$ | $1.51 \pm 0.09$ | $120.2 \pm 15.5$ | $103.7 \pm 5.6$ | 0.25 [11] | 2.00 |
| $SiO_2$ [10] | $1.45 \pm 0.01$ | $1.45 \pm 0.01$ | $0.75 \pm 0.07$ | $-158.2 \pm 6.0$ | $83.8 \pm 1.3$ | 0.17 [12] | 2.17 |

[a]$\kappa$ for 1064 nm is not available.

For a coating with one thin layer, the coefficients in the parenthesis of Eq. (3) are given as [15]

$$\frac{U_B}{F_0^2} = \frac{d_c(1-2\sigma_c)}{3}\left[\frac{Y_c}{Y_s^2}\frac{(1-2\sigma_s)^2(1+\sigma_s)^2}{(1-\sigma_c)^2} + \frac{1}{Y_s}\frac{2(1-2\sigma_s)(1+\sigma_s)(1+\sigma_c)}{(1-\sigma_c)^2} + \frac{1}{Y_c}\frac{(1+\sigma_c)^2}{(1-\sigma_c)^2}\right] \times \frac{1}{\pi w_0^2}, \tag{4}$$

$$\frac{U_S}{F_0^2} = \frac{2d_c}{3}\left[\frac{Y_c}{Y_s^2}\frac{(1-\sigma_c+\sigma_c^2)(1+\sigma_s)^2(1-2\sigma_s)^2}{(1-\sigma_c)^2(1+\sigma_c)} - \frac{1}{Y_s}\frac{(1+\sigma_c)(1-2\sigma_c)(1-2\sigma_s)(1+\sigma_s)}{(1-\sigma_c)^2} + \frac{1}{Y_c}\frac{(1-2\sigma_c)^2(1+\sigma_c)}{(1-\sigma_c)^2}\right]$$

$$\times \frac{1}{\pi w_0^2}, \tag{5}$$

where $d_c$ is the total thickness of the coating. $\sigma_c$ and $\sigma_s$ are the Poisson's ratios and $Y_c$ and $Y_s$ are the Young's moduli of the coating and substrate, respectively. $w_0$ is the laser beam radius at the mirror. For a multilayer coating, the effective Young's modulus $Y_c$ and Poisson ration $\sigma_c$ of the coating can be expressed as [16,17]

$$Y_c = \sum_i Y_i d_i / \sum_i d_i, \tag{6}$$

$$\sigma_c = \sum_i \frac{d_i Y_i \sigma_i}{1-\sigma_i^2} / \sum_i \frac{d_i Y_i}{1-\sigma_i^2}, \tag{7}$$

where $d_i$, $Y_i$, and $\sigma_i$ are the thickness, Young's modulus, and Poisson's ratio of the $i^{\text{th}}$ layer, respectively. Substituting Eqs. (6) and (7) into Eqs. (4) and (5), and subsequently into Eq. (3) together with $\phi_B(f)$ and $\phi_S(f)$, $S_x(f)$ can be obtained. The CTN is defined as the square root of $S_x(f)$. $\phi_B(f)$ and $\phi_S(f)$ can be derived from the measured $\phi(f)$ and the calculated $U_B$, and $U_S$ (see Sec. IV).

A CTN model was previously proposed by Harry et al. [18], in which the elastic energy was divided into components parallel and perpendicular to the coating surface with the associated loss angles $\phi_\parallel$ and $\phi_\perp$, respectively. Both energy components are rigorously not independent and may lead to negative energy in certain situations [15]. Hong's and Harry's models lead to the same expression for thermal noise only when $\phi_B = \phi_S$ and $\phi_\parallel = \phi_\perp$ are assumed in each model. For measurements where the information of $(\phi_B, \phi_S)$ or $(\phi_\parallel, \phi_\perp)$ is insufficient, it is often assumed that the components of the loss angles are equal and both models can be used for a first-order estimation of the thermal noise. The CTNs in Fig. 6 were calculated by using the rigorous Hong's model except in Fig. 6(d), the red solid curve was calculated under the assumption $\phi_B = \phi_S$ as explained in Sec. IV B.

### B. Bulk and shear energy

Strain energy density $u$ of an elastic body is expressed as [19]

$$u = \frac{1}{2}\sigma_{ij}\varepsilon_{ij}, \tag{8}$$

where $\sigma_{ij}$ and $\varepsilon_{ij}$ are the stress and strain tensors. Both tensors are symmetrical under static equilibrium [19,20]. Expansion of $u$ can be expressed as [20]

$$u = \frac{1}{2}(\sigma_{11}\varepsilon_{11} + \sigma_{22}\varepsilon_{22} + \sigma_{33}\varepsilon_{33} + 2\sigma_{12}\varepsilon_{12} + 2\sigma_{13}\varepsilon_{13}$$
$$+ 2\sigma_{23}\varepsilon_{23}). \tag{9}$$

Strain and stress tensors are related by the fourth-rank stiffness tensor $C_{ijkl}$:

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl}. \tag{10}$$

In general, there are only 21 independent elements in $C_{ijkl}$ [21]. For isotropic material such as the amorphous film in our case, only two independent elastic constants remain [21]. Substitute Eq. (10) with the stiffness tensor for the amorphous film, in terms of the bulk modulus $K$ and the shear modulus $\mu$, into Eq. (9), we obtained the energy density for the amorphous film as

$$u = \frac{1}{2}K(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2 + \mu\left[\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2\right.$$
$$\left. + 2(\varepsilon_{12}^2 + \varepsilon_{13}^2 + \varepsilon_{23}^2) - \frac{1}{3}(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2\right]. \tag{11}$$

Integrating the energy density over the volume of interest, we can obtain the total energy $U$:

$$U = U_B + U_S,$$

where

$$U_B = \iiint \frac{1}{2}K(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2 dV, \tag{12}$$

$$U_S = \iiint \mu\left[\varepsilon_{11}^2 + \varepsilon_{22}^2 + \varepsilon_{33}^2 + 2(\varepsilon_{12}^2 + \varepsilon_{13}^2 + \varepsilon_{23}^2)\right.$$
$$\left. - \frac{1}{3}(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33})^2\right]dV. \tag{13}$$

TABLE III. The temperature dependent Young's modulus and Poisson's ratio for common orientations of silicon derived from the measured stiffness coefficients of silicon from [23].

| Temp. (K) | $Y_{[100]}$ (GPa) | $Y_{[110]}$ (GPa) | $Y_{[111]}$ (GPa) | $\sigma_{(111)}$[b] |
|---|---|---|---|---|
| 300 | 130.1 | 169.0 | 187.7 | 0.262 |
| 120 | 131.1 | 170.3 | 189.2 | 0.263 |
| 73 | 131.1 | 170.4 | 189.3 | 0.263 |
| 10[a] | 131.1 | 170.4 | 189.3 | 0.264 |

[a]extrapolated from [23].
[b]on (111) plane of silicon, the Poisson's ratio is isotropic [24].

We used the finite element method (FEM), COMSOL 4.4 [22], to obtain the strain tensor $\varepsilon_{ij}$ for each element, and together with the known elastic constants $K$ and $\mu$ of the film, $U_B$ and $U_S$ can be obtained through the integrations of Eqs. (12) and (13). When using the FEM to calculate the energies, mesh sizes need to be optimized to ensure sufficient convergence of the calculated values under the available computational powers. The convergence test was performed by reducing the mesh sizes in our simulation and the optimal mesh sizes were determined such that variations of the values of $U_B/U$ and $U_S/U$ given in Table V were converged to within $\sim 10^{-4}$ range.

### C. Temperature dependence of elastic constants

The temperature-dependent Young's modulus and Poisson's ratio of the substrate and the film are required in Eqs. (4) and (5) to evaluate the thermal noise for different specifications at different temperature. Table III lists the Young's modulus and Poisson's ratio that we derived from the experimental temperature-dependent stiffness coefficients of silicon [23] for the temperatures of interest. Table III shows that the Young's modulus and Poisson's ratio of silicon are nearly temperature independent within the temperature range of interest. Equations (4) and (5) indicate that a substrate with a larger Young's modulus, as well as a Poisson's ratio that is close to 0.5, is desirable. In terms of this consideration, the (111) silicon is preferred for ET-LF and LIGO Voyager. We do not have measurements of the cryogenic Young's modulus and Poisson's ratio for our films, and here we assume that they are also temperature-independent as the silicon.

## IV. MECHANICAL LOSS

### A. From cantilever ring-down measurement

The mechanical loss was measured by using the cantilever ring-down method. A cantilever was excited at its vibrational resonant frequency $f$ and the decay time $\tau$ of the free-damping was measured. The loss angle of the cantilever $\phi_{\mathrm{can}}(f)$ is expressed as

$$\phi_{\mathrm{can}}(f) = \frac{1}{Q} = \frac{1}{\pi f \tau},$$

where $Q$ is the quality factor of the cantilever mode.

There are two types of cantilever vibrational modes, namely bending mode and torsional mode, as illustrated in Fig. 1 [25].

The loss angles of the bare cantilever substrate $\phi_{\mathrm{sub}}(f)$ and cantilever with coatings $\phi_{\mathrm{coated}}(f)$ are measured and the loss angle of the coating $\phi(f)$ can be derived as

$$\phi(f) = \frac{U_{\mathrm{coated}}}{U}\phi_{\mathrm{coated}}(f) - \frac{U_{\mathrm{sub}}}{U}\phi_{\mathrm{sub}}(f), \qquad (14)$$

where $U_{\mathrm{coated}}$, $U_{\mathrm{sub}}$, and $U$ are the energies of the coated substrate, bare substrate, and the coating, respectively. There are two approaches to solve $\phi(f)$ from Eq. (14) if the elastic constants of the substrate and the film are known. One way is to use the analytical approximations for the energies of the bending modes [26] and the torsional modes [27]. The other way is to use the FEM directly to calculate the energies. In addition to giving an exact solution, the latter approach does not require the calculation of elastic constants for the specific orientations with respect to the crystal axes of the sample. We used the FEM approach to obtain the coating loss angle with the elastic constants of the film given in Table II. The elastic constants of the silicon substrate are given in the built-in data base of the FEM software or can be obtained from Ref. [28].

Cryogenic mechanical losses were measured using the cantilever ring-down method in a cryogenic system [29,30]. The coating loss angles of the single silica film, single silicon nitride film, and 4-pairs and 8-pairs of QW silicon nitride and silica bilayers from 10 K to 130 K are shown in Fig. 2 for two bending modes at 671 Hz and 1,873 Hz and two torsional modes at 1,221 Hz and 3,755 Hz. Cryogenic
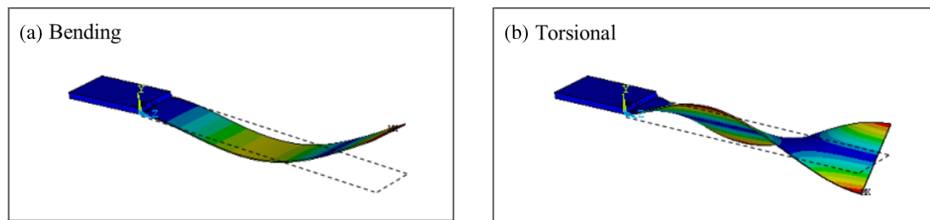


FIG. 1. Illustrations of the vibrational modes of a cantilever in (a) bending mode at 661 Hz and (b) torsional mode at 3642 Hz.
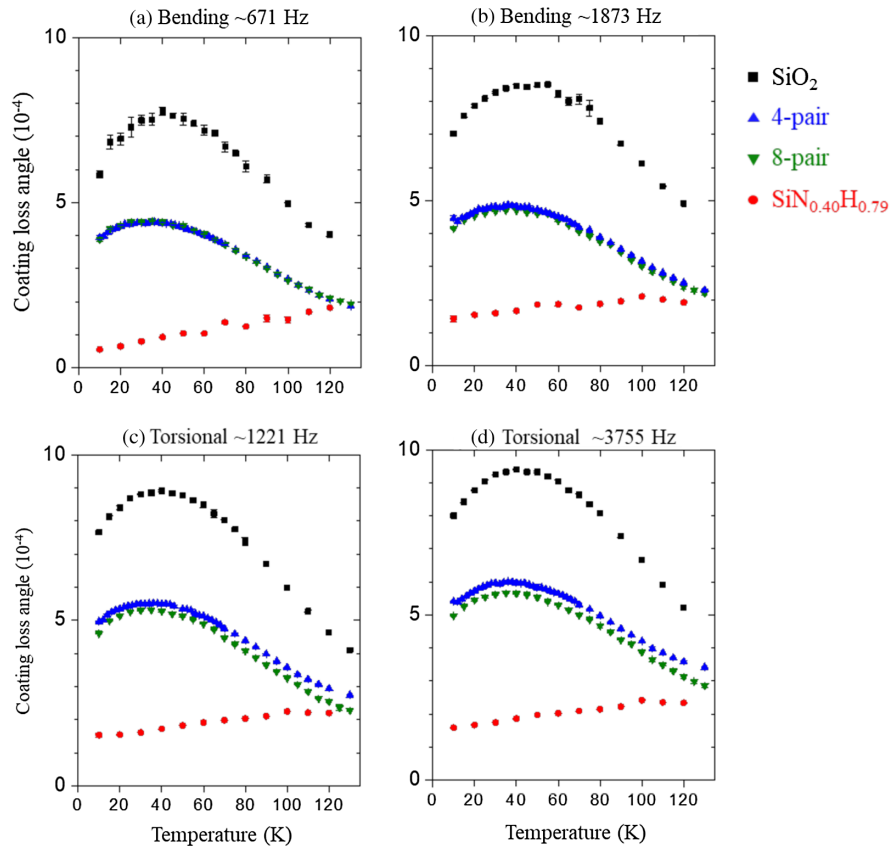
FIG. 2.    Cryogenic loss angles of the single silica film (black square), single silicon nitride film (red dot), and 4-pairs (blue triangle) and 8-pairs (green triangle) of silicon nitride and silica QW bilayers for bending modes at (a) 671 Hz and (b) 1,873 Hz, and torsional modes at (c) 1,221 Hz and (d) 3,755 Hz.

loss measurements for the fundamental mode at approximately 100 Hz are usually skipped owing to the long ring-down duration and the high vibrational noise of the cryogenic system at low frequency and low temperature [29]. However, to measure the loss angle at approximately 100 Hz for notable temperatures such as 10 K (for ET-LF), 20 K (for KAGRA), and 120 K (for LIGO Voyager), we turned off the compressor when the desired temperature was reached and the short decay time ($\approx$120 s) of the thick coated sample, i.e., 8-pair stack, facilitated the ring-down measurement within the time duration when the temperature rise was negligible. Table IV shows the loss angles for the 109-Hz mode, which was a bending mode, for the 8-pair stack at 10 K, 20 K, and 120 K.

The room-temperature mechanical loss was measured by using the cantilever ring-down method in a separate system

from the cryogenic measurement. Each system has its own limitation on measurement [30,31]. The coating loss angles of the 1-,2-,3-,4-, and 8-pairs of QW silicon nitride and silica bilayer samples are shown in Fig. 3. Theoretically, the
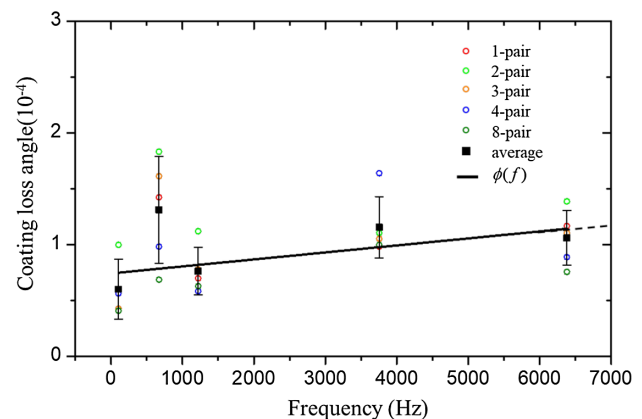


FIG. 3.    Room-temperature loss angles of the 1-,2-,3-,4-, and 8-pairs of silicon nitride and silica QW bilayer samples. The first two modes were bending modes and the rest were torsional modes. The black solid line is the linear fitting of the averages with error bars.

TABLE IV.    Coating loss angles at 10 K, 20 K, and 120 K of the 8-pair silicon nitride and silica QW bilayer for the bending mode at 109 Hz.

| Temperature (K) | 10 | 20 | 120 |
|---|---|---|---|
| $\phi$ at 109 Hz ($10^{-4}$) | $3.9 \pm 0.1$ | $4.4 \pm 0.1$ | $2.0 \pm 0.1$ |

TABLE V.　FEM results for the normalized bulk and shear energies of the coating.

| Mode frequency (Hz) | Mode Type | $U_B/U$ (%) | $U_S/U$ (%) |
|---|---|---|---|
| 101.5 | Bending | 26.84 | 73.16 |
| 657.7 | Bending | 26.78 | 73.22 |
| 1194.2 | Torsional | 1.18 | 98.82 |
| 1843.1 | Bending | 26.88 | 73.12 |
| 3613.3 | Torsional | 2.09 | 97.91 |
| 3634.1 | Bending | 27.06 | 72.94 |
| 5975.6 | Bending | 27.29 | 72.71 |
| 6221.6 | Torsional | 3.75 | 96.25 |

coating loss angle of the alternately deposited thin film stack depends only on the thickness ratio rather than the individual layer thicknesses [32]. Therefore, samples with different numbers of QW pairs should ideally have an equal loss angle. The black squares in Fig. 3 represent the average loss angles of these QW stacks. The first two modes were bending modes and the rest were torsional modes.

## B. Bulk and shear loss angles

Equation (2), restated here, is used to derive the bulk $\phi_B(f)$ and shear $\phi_S(f)$ loss angles from the measured loss angles $\phi(f)$:

$$\phi(f) = \frac{U_B}{U}\phi_B(f) + \frac{U_S}{U}\phi_S(f). \qquad (2)$$

The normalized energies $U_B/U$ and $U_S/U$ were calculated using the FEM method described in Sec. III B with the following inputs: the cantilever has the thickness of 92 $\mu$m and area of 34 mm × 5.5 mm. It has a silicon (001) surface, and the long and short axes of the cantilever are along the [$\bar{1}$10] and the [110] direction respectively. The coating consists of 8 silicon nitride and silica QW bilayers with a total thickness of 3.466 $\mu$m, and an effective Young's

modulus, Poisson's ratio, and density of 91.6 GPa, 0.206, and 2,104 kg/m$^3$, respectively, as derived by using Eqs. (6) and (7) from the values given in Table II.

Table V shows the FEM results of the normalized bulk and shear energies of the coating for each mode and the mode frequency. It is apparent from Table V that the torsional modes are dominated by the shear motion with negligible bulk energies. According to Eq. (2), the shear loss angle at $f_m$, $\phi_S(f_m)$, is therefore approximately equal to the measured loss angle, $\phi(f_m)$, of the torsional mode at $f_m$. $\phi_S(f)$ can therefore be obtained by fitting the $\phi_S(f_m)$ from the torsional modes. The approximation for torsional modes is summarized as

$$\frac{U_B}{U} \ll \frac{U_S}{U}, \qquad \phi_S(f_m) \approx \phi(f_m). \qquad (15)$$

Once $\phi_S(f)$ is known, $\phi_B(f_n)$ from the bending mode at $f_n$ can be obtained from Eq. (2) as

$$\phi_B(f_n) = \frac{U}{U_B}\phi(f_n) - \frac{U_S}{U_B}\phi_S(f_n). \qquad (16)$$

The energy ratios can be obtained from FEM. The results are shown in Figs. 4(a)–4(c) for 10 K, 20 K, and 120 K. Linear fits were applied to the loss angle spectra for both the bulk and shear. Figure 4 reveals that (1) $\phi_B(f)$ is smaller than $\phi_S(f)$, (2) both $\phi_B(f)$ and $\phi_S(f)$ have weak positive frequency dependence, and (3) they are nearly parallel to each other within the range 0 to 5,000 Hz.

For room temperature loss angle, referring to Fig. 3, large uncertainty in the loss angle of the second bending modes leads to $\phi_B(f)$ with an excessively large slope. It is not presently evident how the room-temperature $\phi_B(f)$ would behave in the high-frequency range. Therefore, we assume a single $\phi(f)$, i.e., $\phi_B(f) = \phi_S(f) = \phi(f)$, for the room temperature case as explained in Sec. III A. A linear fit is given to the five measured loss angles as shown in the
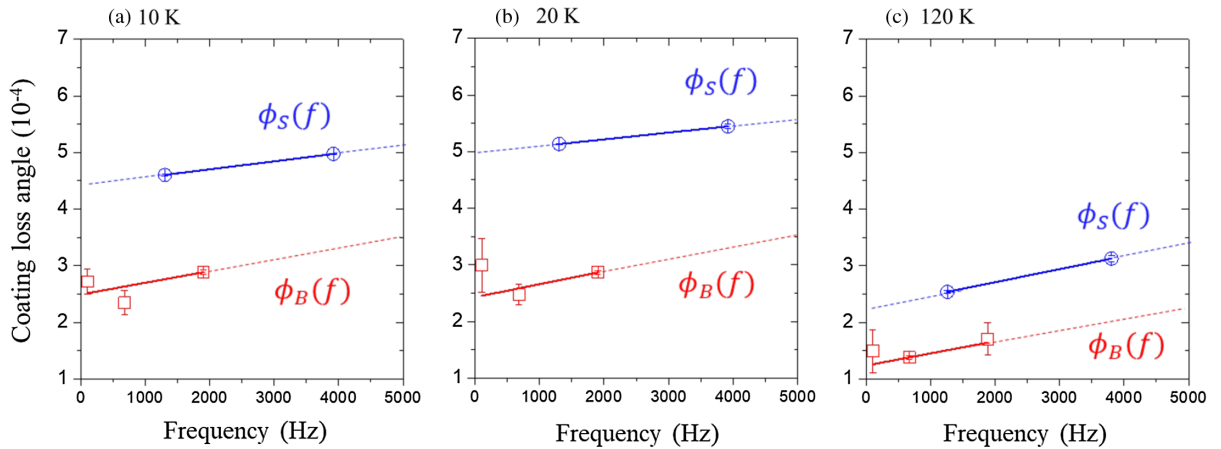


FIG. 4.　Bulk and shear coating loss angles at (a) 10 K, (b) 20 K, and (c) 120 K for the silicon nitride and silica bilayer obtained from the 8-pair sample.

TABLE VI. Specifications of the ETM coating for ET-LF, KAGRA, LIGO Voyager, and aLIGO. T is the transmittance and A is the absorption. $Y_s$ and $\sigma_s$ are the Young's modulus and Poisson's ratio of the substrate, respectively.

| Parameter | ET-LF [3] | KAGRA [5] | Voyager [4,9] | aLIGO [9] |
|---|---|---|---|---|
| Temp. (K) | 10 | 20 | 120 | 290 |
| T (ppm) | 6 | 10 | 5 | 5 |
| A (ppm) | <5 | 0.5–1 | ≤2 | <0.5 |
| $\lambda$ (nm) | 1,550 | 1,064 | 1,550 | 1,064 |
| $w_0$ (cm) | 9.0 | 4.0 | 8.4 | 6.2 |
| CTN[a] | 0.9 | 4.5 | 2.4 | 9.2 |
| Substrate | Silicon | Sapphire | Silicon | Fused silica |
| $Y_s$ (GPa) | 189.3[b] | 400 [5] | 189.2[b] | 72 [35] |
| $\sigma_s$ | 0.26[b] | 0.23 [35] | 0.26[b] | 0.17 [35] |

[a]CTN of the whole detector at 100 Hz with the unit in $10^{-21}$ m/$\sqrt{\text{Hz}}$.
[b]for (111) plane from Table III.

black solid line in Fig. 3 and it is used for the following room temperature CTN evaluation.

## V. COATING THERMAL NOISE EVALUATION

### A. Structures for the QW multilayer high reflectors

The structure of the QW multilayer HR, specifically referred to the coating of the end test mirrors (ETMs), should be determined for different specifications listed in Table VI. Hereafter, "H" represents one QW high-refractive-index layer (silicon nitride) and "L" represents one QW low-refractive-index layer (silica), and the "structure" of the multilayer is symbolized as $\text{Sub}|(\text{LH})^p$, e.g., $\text{FS}|\text{H}(\text{LH})^p$ refers to a structure with one silicon nitride layer (H) adjacent to the fused silica (FS) substrate and p-pairs of silica/silicon nitride $(\text{LH})^P$ on top of it. A design principle to follow is that, for a QW HR stack in air

satisfying the specific transmittance requirement with the least number of layers [33,34], the outermost layer must be H and the innermost layer must be the one with the largest refractive index contrast to the substrate such that the phases of the waves reflected from every interface add up constructively to form HR. There is usually a half-wave thick "optical absentee" protection layer on the top and it is skipped here.

We calculated the transmittance and absorption versus the number of layers for the QW HR stack on a substrate by using the Essential Macleod program [22] with the optical constants of silicon nitride and silica films at 1064 nm and 1550 nm given in Table II. The structures satisfying the transmittance requirement for each specification were determined to be: $\text{Si}|(\text{LH})^{14}$ for ET-LF and LIGO Voyager, $\text{Al}_2\text{O}_3|\text{H}(\text{LH})^{13}$ for KAGRA, and $\text{FS}|\text{H}(\text{LH})^{14}$ for aLIGO. They are shown schematically in Fig. 5 with the relevant



| (a) ET-LF, Voyager | | | (b) KAGRA | | | (c) aLIGO | | |
|---|---|---|---|---|---|---|---|---|

| Structure | Si |(LH)¹⁴ |
|---|---|
| T (ppm) | 3.5 |
| A (ppm) | 45.9 |
| $d_c$ (um) | 6.121 |
| $Y_c$ (GPa) | 91.6 |
| $\sigma_c$ | 0.21 |

| Structure | Al₂O₃ | H(LH)¹³ |
|---|---|
| T (ppm) | 8.6 |
| A (ppm) | NA |
| $d_c$ (um) | 4.004 |
| $Y_c$ (GPa) | 91.9 |
| $\sigma_c$ | 0.21 |

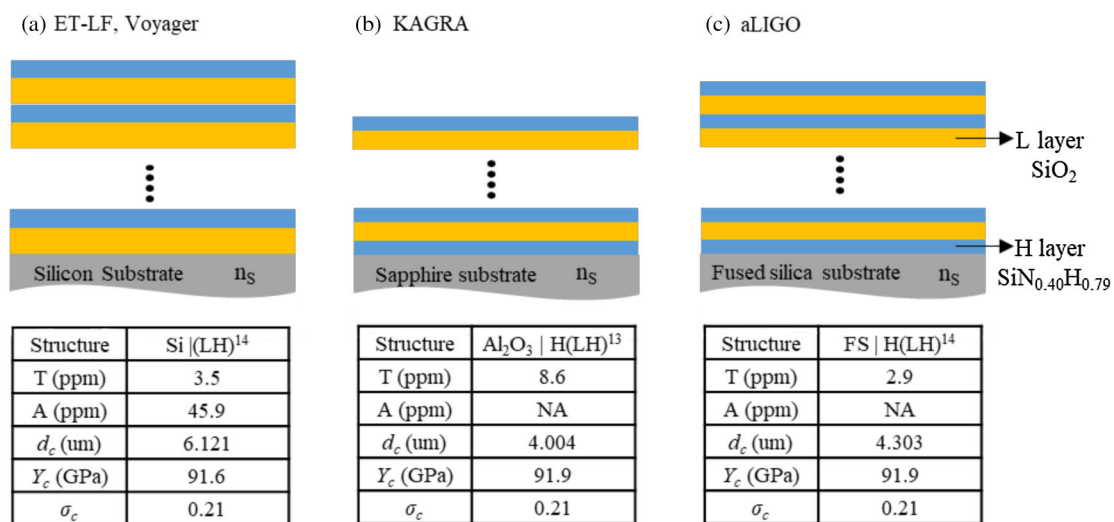| Structure | FS | H(LH)¹⁴ |
|---|---|
| T (ppm) | 2.9 |
| A (ppm) | NA |
| $d_c$ (um) | 4.303 |
| $Y_c$ (GPa) | 91.9 |
| $\sigma_c$ | 0.21 |

FIG. 5. Schematics of the optimal structures for the ETM coating composed of silicon nitride and silica QW HR stack for (a) ET-LF and LIGO Voyager, (b) KAGRA, and (c) aLIGO, where T and A are the optical transmittance and absorption of the stacks, $d_c$ is the total thickness, $Y_c$ and $\sigma_c$ are the effective Young's modulus and Poisson's ratio of coating, respectively.

parameters. The structures of ET-LF and LIGO Voyager have an absorption of 45.9 ppm. Notably, the extinction coefficient used for the calculation, from Table II, was the room-temperature value. For a semiconductor material such as silicon nitride and amorphous silicon, however, the optical absorption at cryogenic temperature is expected to be lower than that at room temperature owing to the reduced inter-band transition at low temperature. Experimental evidence showed that the absorption coefficient of an amorphous silicon film, which is also a potential high-refractive-index material for the mirror coatings of the next-generation laser interferometer gravitational wave detector, at cryogenic temperature was ≈3 times lower than that at room temperature [36]. The thermal-refractive effect, $dn/dT$, is positive for most of the optical materials. Without knowing the quantitative $dn/dT$ for our films, the physical thickness $d_c$ of the film calculated using the room temperature refractive index will be somewhat under-estimated.

## B. Results of thermal noise evaluation

Once the structures are determined, the effective Young's modulus and Poisson's ratio of the coating, $Y_c$ and $\sigma_c$, respectively, can be determined from Table II and Eqs. (6) and (7). The results are listed in the tables of Fig. 5 together with the total coating thickness $d_c$. Substituting all the

relevant parameters into Eqs. (4) and (5), $U_B/F_0^2$ and $U_S/F_0^2$ can be obtained. Substituting $U_B/F_0^2$, $U_S/F_0^2$ together with $\phi_B(f)$ and $\phi_S(f)$ from Fig. 4 and 3 into Eq. (3), CTN of the ETM coatings can thus be obtained. The results are shown in Figs. 6(a)–6(d) as the solid curves.

CTN of the whole detector includes the contributions from the two ETMs and the two input test mirrors (ITMs). It equals the square root of the sum of two times square of the CTN of the ETM and ITM [37] for uncorrelated thermal noise. ITMs have lower transmittance than ETMs and, therefore, should have less number of layers and, hence, lower CTN. Without knowing the detail coating structures of the ITMs, the CTN of the ETM coating is equal to or greater than one half of the CTN of the whole detector. The lower limits of CTN specification for the coatings of the ETM for ET, KAGRA, LIGO Voyager, and aLIGO are shown in Figs. 6(a)–6(d) as the dash curves. The directly measured CTN of the ETM for the current coating of aLIGO is shown in Fig. 6(d) as the orange dot curve [38]. CTN of the ETM for aLIGO calculated using the coating parameters in Table V of Ref. [39], where a single $\phi$ was assumed for each film, is shown in Fig. 6(d) as the black dot curve.

Figure 6 shows that, at 100 Hz, the CTN of the silicon nitride and silica QW HR is 1.8 times higher than the lower
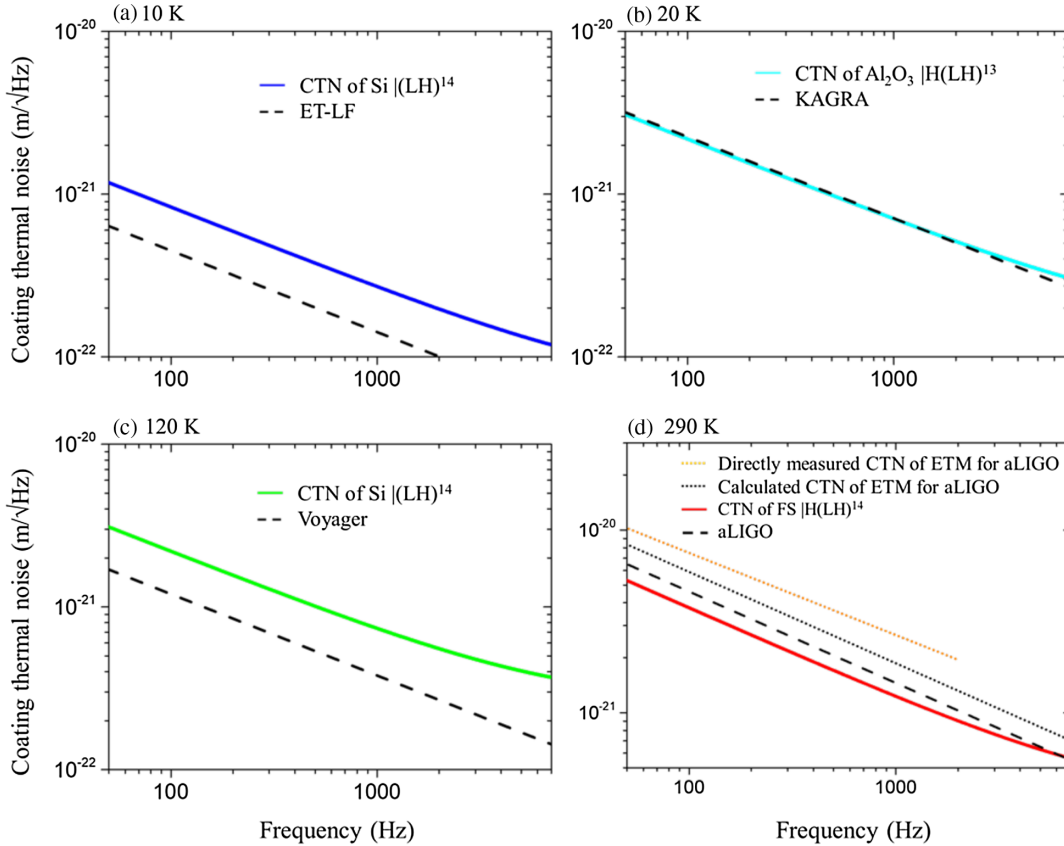


FIG. 6. Comparison of the calculated CTN of the silicon nitride and silica QW HR (solid curves) with the lower limit of CTN of the ETM (dash curves) for (a) ET-LF, (b) KAGRA, (c) LIGO Voyager, and (d) aLIGO.

TABLE VII.  Summary of the optical transmittance, absorption, and CTN of silicon nitride and silica QW HR and comparison with the specifications of ETM in ET-LF, KAGRA, LIGO Voyager, and aLIGO.

| ETM properties | ET-LF | | KAGRA | | Voyager | | aLIGO | |
|---|---|---|---|---|---|---|---|---|
| | Spec. [3] | Si\|(LH)[14] | Spec. [5] | $Al_2O_3$ \|H(LH)[13] | Spec. [4] | Si\|(LH)[14] | Spec. [9] | FS\|H(LH)[14] |
| T (ppm) | 6 | 3.5 | 10 | 8.6 | 5 | 3.5 | 5 | 2.9 |
| A (ppm) | <5 | 45.9 | 0.5–1 | NA | ≤2 | 45.9 | <0.5 | NA |
| CTN[a] | 0.45[b] | 0.83 | 2.25[b] | 2.18 | 1.20[b] | 2.20 | 7.50[c] | 3.75 |

[a]frequency at 100 Hz with the unit in $10^{-21}$ m/$\sqrt{Hz}$.
[b]lower limit for CTN of the ETM.
[c]directly measured CTN of the ETM [38].

limit of the ETM of ET-LF, approximately equal to that of KAGRA, 1.8 times higher than that of LIGO Voyager, and 2 times lower than that of the directly measured CTN of the ETM of aLIGO. Table VII summarizes the transmittance, absorption, and CTN of the silicon nitride and silica QW HR in comparison with the specifications of ET, KAGRA, LIGO Voyager, and aLIGO.

Both silicon nitride and silica were not subjected to post-deposition heat treatment. It has been shown that the mechanical loss angle of the single silica film can be reduced by varying the deposition temperature and performing post-deposition heat treatment [40]; compatible heat treatment for silicon nitride with silica shall be explored to reduce the mechanical loss of the bilayer and consequently the CTN of the QW HR. Postdeposition heat treatment was also shown to be an effective means to reduce the optical absorption of IBS amorphous silicon [41], and it would be advantageous to explore the effect of thermal annealing on the optical absorption of silicon nitride and silica. The CTN for the current silicon nitride and silica QW HR with respect to the specifications of ET-LF and LIGO Voyager could be improved by implementing the optimized none-QW thickness combination of the

silicon nitride and silica layers to minimize the CTN through Eqs. (3)–(7) and maint the required transmittance [42,43].

Figures 4 and 3 reveal that all the loss angles of the silicon nitride and silica bilayer have a weakly positive linear frequency dependence within the frequency range of interest as follows:

$$\phi(f) = a + bf, \qquad a \gg b > 0. \qquad (17)$$

Using the case of room-temperature loss angle to elaborate this effect, we determined that $a$ is $7.41 \times 10^{-5}$ and $b$ is $6.20 \times 10^{-9}$ at room temperature from Fig. 3. Substituting Eq. (17) into Eq. (3), it can be shown that the CTN deviates from its normal $1/\sqrt{f}$ dependence and bends asymptotically toward $\sqrt{b \times C}$, where $C = 4K_B TU/\pi F_0^2$, as illustrated in Fig. 7, with the bend-over frequency defined as $a/b$. It is evident that a lower loss angle and higher bend-over frequency are desirable in general; i.e., less-dispersive materials with lower loss angle are preferred. For silicon nitride and silica bilayer, the bend-over frequency is 11 962 Hz at room temperature. The coefficients $a$ and $b$ for IBS $Ta_2O_5$ and IBS silica were presented in Ref. [44]. It can be shown using Eq. (18) that the bend-over frequency for IBS $Ta_2O_5$/silica is 174 382 Hz at room temperature. Direct measurement of the CTN for the current coatings of aLIGO [38], IBS Ti:$Ta_2O_5$/silica, within the frequency range 40 Hz to 2000 Hz showed a slope of $-0.45$ rather than $-0.5$ for log CTN vs log $f$, which also indicates the weakly positive frequency dependence of the loss angle, possibly from the coating materials.

## VI. MULTIMATERIAL STACK FOR IMPROVEMENT OF OPTICAL ABSORPTION

Since the standing wave pattern for a QW HR stack peaks at the interfaces of the front few layers (the layers facing the incident beam) and drops rapidly toward the back layers [45], referring to Fig. 8, it was proposed and simulated [46,47] that, for an HL material system with superior mechanical loss but inferior optical absorption such as amorphous silicon and silica [47] and our silicon
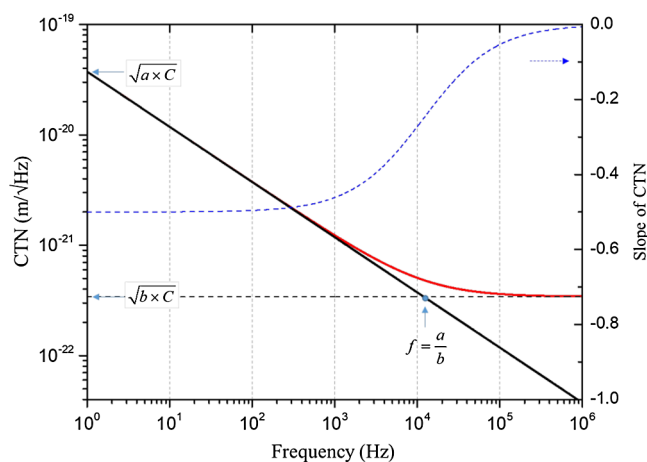
FIG. 7.  CTN for silicon nitride and silica QW HR coating with (red solid curve) and without (black solid curve) a linear frequency dependence of the loss angle $\phi(f) = a + bf$. The blue dash curve is the slope of the red curve. $C = 4K_B TU/\pi F_0^2$.
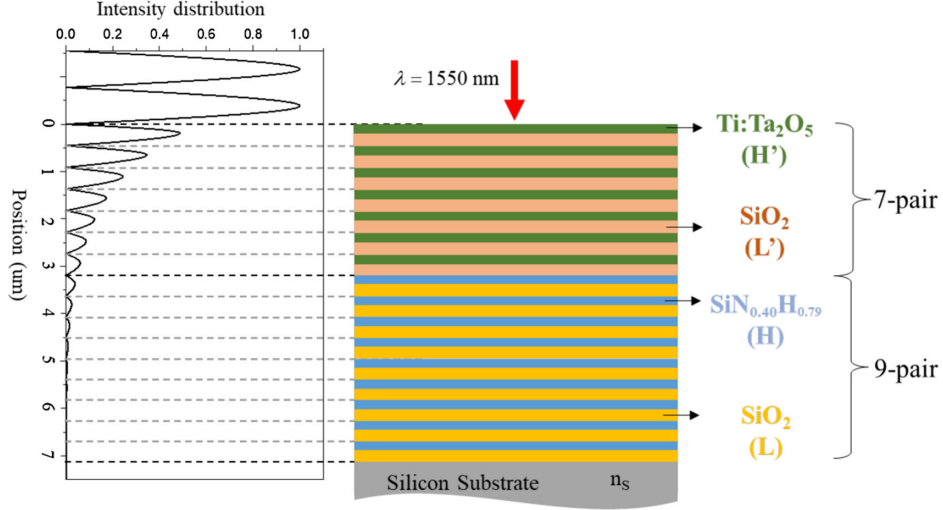
FIG. 8.    Multimaterial structure. Each layer has QW optical thickness at the wavelength 1550 nm and the total thickness $d_c$ is 7.129 um.

nitride and silica, the HL stack could be coupled to another stack composed of HL materials with superior optical absorption but inferior mechanical loss, such as the conventional Ti : Ta$_2$O$_5$/silica for aLIGO, in a way that the former is deposited in the back of the HR and the latter is deposited in the front of the HR; the end result is a composite HR stack, referred to as a "multimaterial system," with reduced optical absorption and yet reasonable mechanical loss.

The following optimization for the multimaterial system was specifically aimed for a silicon substrate and the wavelength of 1550 nm, i.e., for ET-LF and LIGO Voyager. In the simulation, we started with the Si|(silica/silicon nitride)$^{14}$ PECVD QW HR, referred to as Si|(LH)$^{14}$, of the structure in Fig. 5(a). QW pairs of IBS (silica/Ti : Ta$_2$O$_5$) referred to as (L′H′), with the optical constants listed in Table VIII, were added onto the Si|(LH)$^{14}$ pair by pair. Optical absorption was calculated for the whole stack after adding each pair. The addition of (L′H′) pairs was stopped when the optical absorption reduced to below 2 ppm (the specification of LIGO Voyager). The structure formed up to this point was Si|(LH)$^{14}$(L′H′)$^7$. The (LH) pairs were thereafter removed pair by pair, and the transmittance of the whole stack was calculated after the removal of each pair. The removal was stopped when the transmittance increased to no more than 5 ppm. The final structure formed through this process was Si|(LH)$^9$(L′H′)$^7$ as shown in Fig. 8 schematically. The

transmittance was 2.5 ppm and the absorption was 2 ppm. The CTN of the final structure is calculated as follows.

The Young's moduli, Poisson's ratios, and loss angles of the IBS silica and IBS Ti : Ta$_2$O$_5$ were reported in Ref. [49,50,53] and are summarized in Table VIII. Since the loss angles of IBS Ti : Ta$_2$O$_5$ and IBS silica from Ref. [50,53] were not distinguished between the bulk and shear, the CTN for the Si|(LH)$^9$(L′H′)$^7$ should be calculated by using the CTN model with a single $\phi$.

The effective loss angle of (LH)$^9$(L′H′)$^7$, $\phi$, can be obtained by substituting the loss angle, Young's modulus, and thickness of the individual layer from Tables II, IV, and VIII into Eq. (18) given below [32]:

$$\phi = \sum_i \phi_i Y_i d_i / \sum_i Y_i d_i. \qquad (18)$$

The effective Young's modulus $Y_c$ and Poisson's ratio $\sigma_c$ of (LH)$^9$(L′H′)$^7$ can be obtained by the same method described in Section V B. Substituting $\phi$, $Y_c$, $\sigma_c$, and the total thickness $d_c$ of (LH)$^9$(L′H′)$^7$ into Eqs. (3)–(5) by assuming $\phi_B(f) = \phi_S(f) = \phi$, the CTN of Si|(LH)$^9$(L′H′)$^7$ multimaterial QW HR on a silicon substrate at the wavelength 1550 nm was obtained. The optical transmittance, absorption, and CTN of silicon nitride and silica QW HR and multimaterial QW HR, and the corresponding specifications of ET-LF and LIGO Voyager are summarized and listed in Table IX for comparison.

TABLE VIII.    Material properties of IBS Ti : Ta$_2$O$_5$ and silica used for multimaterial analysis.

| Materials | $n$ (1550 nm) | $\kappa$ (1550 nm) ($10^{-7}$) | $Y$ (GPa) | $\Sigma$ | $\phi$ ($10^{-4}$) 10 K | 120 K |
|---|---|---|---|---|---|---|
| Ti : Ta$_2$O$_5$[a] | 2.05 [48] | 5.5[b] [47] | 140 [49] | 0.23 [49] | 4.6 [50] | 3.2 [50] |
| SiO$_2$[a] | 1.45 [49] | Negligible [51,52] | 72 [49] | 0.17 [49] | 7 [53] | 1.7 [53] |

[a]deposited via IBS and annealed at 600 °C for 24 h.
[b]estimated using the HR absorption in [47] and assuming $\kappa$ of silica is negligible.

TABLE IX. Comparison of the transmittance, absorption, and CTN between silicon nitride and silica QW HR, multimaterial QW HR, and the specifications of ET-LF and LIGO Voyager.

| ETM properties | ET-LF | | | Voyager | | |
|---|---|---|---|---|---|---|
| | Spec. [3] | Si$\|$(LH)$^{14}$ | Si$\|$(LH)$^9$(L$'$H$'$)$^7$ | Spec. [4] | Si$\|$(LH)$^{14}$ | Si$\|$(LH)$^9$(L$'$H$'$)$^7$ |
| T (ppm) | 6 | 3.5 | 2.5 | 5 | 3.5 | 2.5 |
| A (ppm) | <5 | 45.9 | 2.0 | ≤2 | 45.9 | 2.0 |
| CTN[a] | 0.45[b] | 0.83 | 1.09 | 1.20[b] | 2.20 | 2.81 |

[a]frequency at 100 Hz with the unit in $10^{-21}$ m/$\sqrt{\text{Hz}}$.
[b]lower limit of CTN of the ETM.

Table IX shows that, for the multimaterial Si$\|$(LH)$^9 \times$ (L$'$H$'$)$^7$, the absorption reduced significantly from 45.9 ppm to 2 ppm and the transmittance remained within the specification. The total thickness of Si$\|$(LH)$^9$(L$'$H$'$)$^7$ increased to 7.13 $\mu$m as compared with that (6.12 $\mu$m) of Si$\|$(LH)$^{14}$, and the CTN at 100 Hz increased about 31% for ET-LF and 28% for LIGO Voyager. A trade-off between the optical absorption and CTN by adjusting the pair numbers of (LH) and (L$'$H$'$) is possible if the specifications are changed. In general, more (L$'$H$'$) pairs will give lower optical absorption but higher CTN.

To realize multimaterial coatings, the deposition sequence of Si$\|$(LH)$^9$(L$'$H$'$)$^7$ is designed as follows: (silicon nitride/silica)$^9$ is deposited continuously in the PECVD chamber on a silicon substrate first and is thereafter moved to the IBS chamber to deposit the (Ti:Ta$_2$O$_5$/silica)$^7$ stack. Since the optical and mechanical properties of the Ti:Ta$_2$O$_5$/silica QW stack were optimized via postdeposition annealing at 600 °C it is expected that the PECVD silicon nitride and silica QW stack should be able to sustain annealing at 600 °C without degradation. It was reported that the amorphous PECVD silicon nitride could sustain annealing up to 800 °C without crystallization [54]. It remains to be seen if the annealing performed at 600 °C on the PECVD silicon nitride and silica stack would improve the characteristics of silicon nitride in terms of absorption and mechanical loss angle as it had improved in the case of amorphous silicon [36,55].

## VII. CONCLUSION

In this report, the theories and methods for calculating the thermal noise using Hong's model with bulk and shear loss angles derived from the cantilever ring-down measurement for silicon nitride and silica QW coatings were demonstrated in detail. We have shown that the coating thermal noise of a QW HR composed of silicon nitride and silica bilayer deposited via the PECVD method are within two times of the lower limit of the ETM for ET-LF and LIGO Voyager and about equal to that of KAGRA. The coatings also showed lower coating thermal noise than that of the current room-temperature detector of aLIGO. The loss angle of the materials showed a linear weakly positive frequency dependence. The optical absorption of the as-deposited silicon nitride and silica QW HR is 45.9 ppm at 1550 nm, which is higher than expected. However, the optical absorption of a multimaterial structure composed of seven pairs of IBS Ti:Ta$_2$O$_5$/silica and nine pairs of PECVD silicon nitride and silica on a silicon substrate can significantly be reduced to 2 ppm at 1550 nm, which satisfies the specifications of ET-LF and LIGO Voyager. Thermal annealing and optimized layer design are anticipated to further reduce the thermal noise and optical absorption of the silicon nitride and silica HR.

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