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Mirror Coating Solution for the Cryogenic Einstein Telescope

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(Received 11 December 2018; published 13 June 2019)

Planned cryogenic gravitational-wave detectors will require improved coatings with a strain thermal noise reduced by a factor of 25 compared to Advanced LIGO. We present investigations of HfO_2 doped with SiO_2 as a new coating material for future detectors. Our measurements show an extinction coefficient of $k = 6 \times 10^{-6}$ and a mechanical loss of $\phi = 3.8 \times 10^{-4}$ at 10 K, which is a factor of 2 below that of SiO_2, the currently used low refractive-index coating material. These properties make HfO_2 doped with SiO_2 ideally suited as a low-index partner material for use with *a*-Si in the lower part of a multimaterial coating. Based on these results, we present a multimaterial coating design which, for the first time, can simultaneously meet the strict requirements on optical absorption and thermal noise of the cryogenic Einstein Telescope.

DOI: 10.1103/PhysRevLett.122.231102

Introduction.—During the first two observing periods of advanced interferometric gravitational-wave detectors, ten gravitational-wave signals from binary black hole mergers and one from a binary neutron star inspiral have been measured [1–6]. To improve upon the sensitivity of the current generation of detectors, Advanced LIGO [7,8] and Advanced Virgo [9], it is essential to reduce coating thermal noise (CTN). The CTN amplitude spectral density is proportional to the square root of the mirror temperature [10]. Therefore, gravitational-wave detectors such as KAGRA [11,12] and the low frequency detector of the planned Einstein Telescope (ET-LF) [13] will operate at low temperatures. At frequencies of around 10 Hz, ET-LF will be 100 times more sensitive than Advanced LIGO and Virgo at the same frequency. This improved sensitivity will increase the observable volume of space by a factor of 100^3 and open up the 1–10 Hz frequency band. This may allow multiple detections of known young pulsars [14], first detections of a Galactic type Ia supernova [15], and many distant—and possibly new types of—sources. The expansion of the frequency range will also allow inspirals to be observed for a longer time before the final merger events.

The interferometer mirror coatings are made of alternating layers of materials with low and high refractive index n. In the simplest case, the layers are a quarter of a wavelength (QWL) in optical thickness (n multiplied by the geometric thickness t). To avoid thermal deformation of the mirrors and to maintain the desired cryogenic temperature, heating must be minimized. Therefore, in addition to low CTN, low optical absorption at the ppm (10^{-6}) level is required.

SiO_2 and Ta_2O_5 (or Ta_2O_5 doped with TiO_2 [16]), deposited using ion-beam sputtering (IBS), are widely used coating materials with very low absorption and scattering [17]. A complication of cooling is that CTN is proportional to the square root of the mechanical loss, which is temperature dependent. Both SiO_2 and Ta_2O_5 (doped or undoped) show mechanical-loss peaks at low temperatures [18–20]. There is some uncertainty if these peaks are present in multilayer coatings formed from these materials [21,22]. However, it is clear that the mechanical loss is too high to meet the sensitivity requirements of ET-LF.

Another complication is that fused silica, the currently used mirror substrate material, is not suitable for low temperature operation due to a large peak in mechanical loss at around 40 K [23–25]. For ET-LF, the use of crystalline silicon (*c*-Si) is planned [13]—the material is also used for the mechanical spacer (at 124 K) in stable reference cavities for optical frequency standards [26]. *c*-Si is not transparent at 1064 nm. Therefore, a change to a longer laser wavelength is required [27], with 1550 nm planned for ET-LF.

Amorphous silicon (*a*-Si) is a very interesting coating material due to low mechanical loss at low temperatures [28,29]. Currently, the best estimated absorption for a highly reflective multilayer *a*-Si/SiO_2 coating is 7.6 ppm at 1550 nm and room temperature ($k_{a-\mathrm{Si}} = 1.22 \times 10^{-5}$) [30]. There is also potential for further reduction at a higher wavelength and a lower temperature [31,32]. To obtain the minimum optical absorption in *a*-Si, heat treatment at 400 °C is required. Thus a low-index partner material also must have good optical properties and mechanical loss at this heat-treatment temperature.

Using a-Si (instead of Ta_2O_5) in a highly reflective coating with SiO_2 would significantly decrease CTN at low temperatures. However, this decrease is limited by the mechanical loss of the SiO_2 layers. To meet the ET-LF requirements, it is therefore essential to find an alternative low-index material for combination with a-Si.

This Letter presents IBS HfO_2 doped with SiO_2 ($SiO_2:HfO_2$) as a low-index material for ET-LF coatings. HfO_2 films have been observed to be partially polycrystalline, with the degree of crystallinity increasing upon heat treatment. This polycrystalline structure causes a problematically high level of optical scattering [33]. However, HfO_2 shows lower mechanical loss [33] than SiO_2. Doping HfO_2 with SiO_2 has been shown to stabilize the coating against crystallization following heat treatment at temperatures up to 550 °C [34,35]. We show that $SiO_2:HfO_2$ used with a-Si can meet the optical absorption requirements (<5 ppm) and the CTN requirements of ET-LF at an operating temperature of 10 K [13] when used together with SiO_2 and Ta_2O_5 in a multimaterial design [36,37].

Deposition and heat treatment.—Coating mechanical loss was measured with a ringdown technique as described in Ref. [18] using cantilevers coated with a HfO_2 layer doped with 27% SiO_2 (measured by x-ray photoelectron spectroscopy). The coatings were deposited by CSIRO [38] using IBS. Ellipsometry was used to estimate the thickness of the as-deposited coating to be (483 ± 3) nm. The cantilevers were made of c-Si, which has low mechanical loss below 150 K [39,40], to maximize the sensitivity to the coating loss. Prior to coating deposition, an oxide layer (SiO_2) was grown on the cantilevers by thermal oxidation, to ensure good adhesion of the coating. The oxide layer was approximately 20 nm thick, which was also measured via ellipsometry.

Optical coatings are commonly heat treated to reduce the stress and optical absorption [41]. Coating mechanical loss is also often strongly dependent on heat treatment [19]. Therefore, the coated cantilevers were heat treated for 24 h at temperatures of 150 °C, 300 °C, 400 °C, and 600 °C by CSIRO to cover the typical temperature span used by commercial vendors. There is some evidence in the literature of the growth of a few nanometers of oxide due to heat treatment for HfO_2 films on c-Si [42], although it should be noted that this is predicted to occur at higher temperatures than are used here. Our ellipsometry measurements showed no significant variation in thickness of the SiO_2-doped HfO_2 coating due heat treatment. For the oxide layer, there was no evidence of a significant increase in thickness after heat treatment at 400 °C—the temperature used for the mechanical-loss results presented here. For heat treatment at 600 °C, a maximum possible increase in oxide thickness of 6 nm was estimated. It should be noted that variations of up to 3 nm were observed for samples with identical heat treatment.

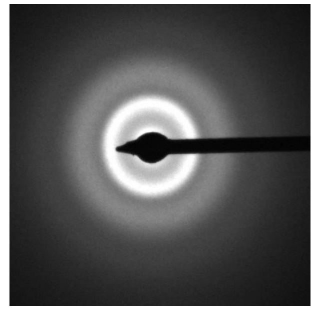

FIG. 1. Electron diffraction pattern of the 600 °C heat-treated silica-doped hafnia coating showing the coating to still be amorphous. This pattern is representative of those measured at lower heat-treatment temperatures.

Transmission electron microscope measurements of coatings deposited on SiO_2 substrates indicated that all of the heat-treated coatings remained amorphous (see Fig. 1). This keeps optical scattering low and makes SiO_2 ($SiO_2:HfO_2$) potentially useful as a coating material for gravitational-wave detectors.

Mechanical loss and Young's modulus.—The Young's modulus, Y, of the coating is required for calculation of the coating mechanical loss [43]. For $SiO_2:HfO_2$, $Y = 180$ GPa was calculated [44] using the moduli of both SiO_2 and HfO_2 (see Table I).

The mechanical losses of several bending modes in the frequency range 0.5 to 9.5 kHz were measured between 10 and 200 K. After a complete measurement cycle, the cantilever was reclamped and the measurements repeated. This ensures that unintentional variations in the clamping procedure did not affect the results. The mechanical loss of the coatings was calculated by comparing the mechanical loss of the coated c-Si cantilevers with nominally identical oxidized, uncoated samples using Ref. [43]. Underestimating the oxide thickness of the heat-treated, coated samples would result in a small overestimation of the

TABLE I. Material properties used for CTN calculations. The heat-treatment temperature for the losses (ϕ) was 450 °C for SiO_2 and 400 °C for all other materials, with loss values at 600 °C in brackets.

Material	$\phi(\times 10^{-4})$ 10 K	n	k $(\times 10^{-5})$	Y (GPa)
SiO_2	8.5 (5) [45]	1.44 [46]	0.008[a]	72 [47]
HfO_2				220 [48]
$SiO_2:HfO_2$	3.8 ± 0.3	1.91 [49]	0.40 ± 0.09	180 [49]
Ta_2O_5	5 (7) [19]	2.05 [50]	0.008[a]	140 [47]
a-Si	≤ 0.17[b] [30]	3.48 [51]	1.22 ± 0.21 [30]	147 [48]

[a]Effective k chosen for $\alpha_{HR} \leq 0.5$ ppm. This assumes that the effective k value for the stack at 1550 nm is identical to 1064 nm [52] so that the absorption scales just with layer thickness.
[b]Measured only measured at room temperature.

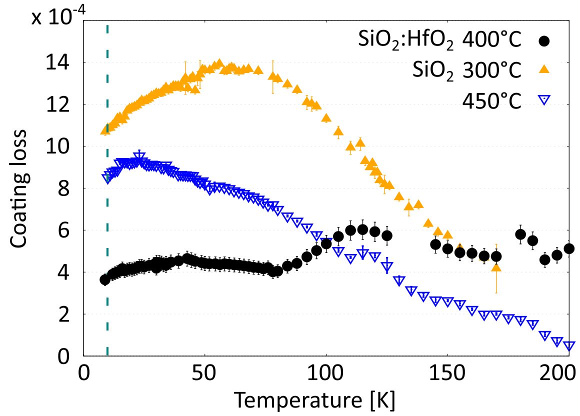

FIG. 2. Temperature dependent coating mechanical loss of $SiO_2:HfO_2$ heat treated at 400 °C (black circles) measured on a resonant mode at 1.4 kHz. Also shown is the mechanical loss of an IBS SiO_2 coating at different heat-treatment temperatures [45]. The dashed vertical line marks a temperature of 10 K.

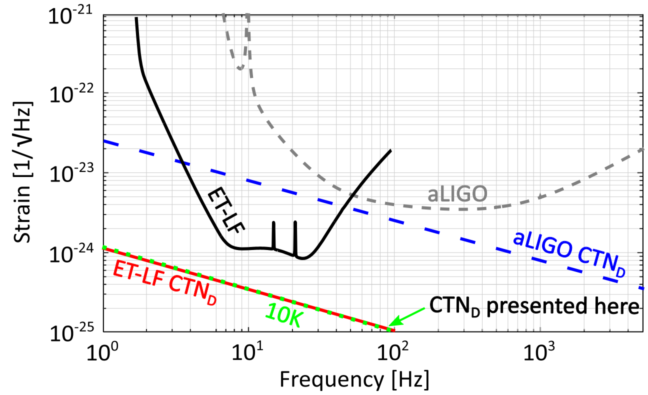

FIG. 3. Design sensitivity (gray dashed curve) and CTN_D (blue dashed line) of Advanced LIGO and design sensitivity (black curve) and CTN_D (red line) of ET-LF. The green dotted line shows CTN_D of our coating [coating (c) in Table II] at a mirror temperature of 10 K.

coating loss. For 400 °C heat treatment, there was no evidence of oxide growth. (For the possible 6 nm oxide growth at 600 °C, the coating loss would change by ≈1%.)

Good agreement was obtained between the measured coating loss for each bending mode. Figure 2 shows a representative data series at a mode frequency of 1.4 kHz. The data shown are for heat treatment at 400 °C which is the optimum temperature for minimizing the absorption in the high-index *a*-Si layers in a highly reflective coating stack.

Below 40 K, the loss of the $SiO_2:HfO_2$ heat treated at 400 °C is significantly lower than the loss of IBS SiO_2 (heat treated at 300 °C and 450 °C), as shown in Fig. 2. $SiO_2:HfO_2$ heat treated at 400 °C therefore has great potential as a low thermal-noise replacement for SiO_2 coating layers.

Optical absorption.—Fused silica disks were coated with $SiO_2:HfO_2$ in the same coating run as the cantilevers used for mechanical-loss studies. The absorption of the coatings was measured at 1550 nm using photothermal common-path interferometry [53]—a technique based on measuring a thermal effect due to optical absorption. The absorption of the as-deposited coating was found to be (25 ± 5) ppm for a 500 nm thick layer. The error originates from variations in absorption across the sample and from reproducibility after realignment. This absorption corresponds to an extinction coefficient of $k = (6.4 \pm 1.3) \times 10^{-6}$. The absorption coefficient of a coating layer, α, is related to the extinction coefficient, k, by $\alpha = 4\pi k/\lambda$. The total absorption of an highly reflective (HR) coating, α_{HR}, also includes the effect of interference in the layers. After heat treatment at 400 °C, which is the optimum temperature for mechanical loss, the absorption reduces to (16 ± 3) ppm $[k = (4.0 \pm 0.9) \times 10^{-6}]$.

Discussion.—Figure 3 shows the total strain noise of the Advanced LIGO detectors (gray dashed curve) at their design sensitivity. The black solid curve represents the total strain noise of the ET-LF design [13]. This strain noise can be converted into displacement noise by multiplying by the detector arm length (4 km for aLIGO, 10 km for ET-LF), allowing comparison between detectors to be unbiased by differing arm lengths. The coating displacement thermal noise of the whole detector, CTN_D, includes contributions from the two input test masses (ITMs) and the two end test masses (ETMs) forming the interferometer arm cavities:

$$CTN_D = (2 \times CTN_{ETM}^2 + 2 \times CTN_{ITM}^2)^{1/2}. \quad (1)$$

The CTN_D requirement for ET-LF is $\approx 3.6 \times 10^{-21}\ \mathrm{m}/\sqrt{\mathrm{Hz}}$ at a reference frequency of 10 Hz (shown in terms of strain noise by the red solid line)—this is about a factor of 25 below the CTN_D of Advanced LIGO (blue dashed line) [7].

The Einstein Telescope design study suggests an operation temperature of 10 K, with the optical absorption of the coating required to be ≤ 5 ppm [13]. The design transmission of the ETMs is T ≈ 6 ppm, and of the ITMs T ≈ 7000 ppm [13]. For the coating materials used in current gravitational-wave detectors, SiO_2 and Ta_2O_5, CTN_D would be $\approx 6.45 \times 10^{-21}\ \mathrm{m}/\sqrt{\mathrm{Hz}}$ at 10 Hz and 10 K [see Table II(a)], calculated using Ref. [10]. Table II also shows CTN for the ETMs and ITMs separately. For the ITMs, CTN is lower, as fewer layers are required to provide the lower design reflectivity.

Coating (b) in Table II demonstrates the potential of using $SiO_2:HfO_2$ as a low-index material alongside *a*-Si. Based on the results presented here, this combination of materials results in a $CTN_D = 2.4 \times 10^{-21}\ \mathrm{m}/\sqrt{\mathrm{Hz}}$ at 10 K. This surpasses the requirement for ET-LF. However, the absorption of this coating, of $\approx(11.9 \pm 2.3)$ ppm at 1550 nm, exceeds the required value by more than a factor of 2.

A way to further reduce the absorption is the use of a multimaterial design [36,37]. In this design, a few

TABLE II. CTN of different coatings on c-Si substrates at a reference frequency of 10 Hz, a temperature of 10 K and a beam radius of 9 cm. The material parameters used are shown in Table I.

Case	Bilayers ETM (ITM)	Transmission ETM (ITM) (ppm)	Heat treatment (°C)	CTN ETM (ITM) ($\times 10^{-21}$ m/$\sqrt{\text{Hz}}$)	CTN_D	α_{HR} (ppm)
(a)	$18\,(7) \times SiO_2/Ta_2O_5$	4 (8500)	600	4.0 (2.4)	6.6	0.6
(b)	$10\,(4) \times SiO_2{:}HfO_2/a\text{-Si}$	2 (9000)	400	1.4 (0.9)	2.4	11.9
(c)	$2 \times SiO_2/Ta_2O_5 + 10\,(4) \times SiO_2{:}HfO_2/a\text{-Si}$	4.4 (6000)	400	1.9 (1.6)	3.5	3.4
ET-LF requirement [13]		5 (7000)			≈3.6	≤ 5

low-absorbing layers are used on top of the coating to reduce the laser power reaching the lower, higher-absorbing layers. In our case, two bilayers of SiO_2 and Ta_2O_5 reduce the light intensity enough for the absorption to be within the ET-LF requirement. This absorption reduction comes at the expense of a slight increase in CTN_D, which still meets the requirement [3.6×10^{-21} m/$\sqrt{\text{Hz}}$ at 10 K; see Table II(c)]. The exact layer design and the light intensity inside the coatings is shown in Fig. 4(a) for the ETMs, and in Fig. 4(b) for the ITMs. The thickness of the layer of $SiO_2{:}HfO_2$ closest to the substrate has been adjusted to be 0.2 QWL thick, allowing the transmission requirement for the ET-LF ITM mirror to be matched more closely. This coating design therefore meets the ET-LF requirements on thermal noise and optical absorption. The total CTN_D strain noise for these coatings is shown by the green dotted line in Fig. 3. For this coating, heat treatment at 400 °C was assumed to minimize the optical absorption of the a-Si layers, which increases the mechanical loss of SiO_2 and Ta_2O_5 compared to coating (a) (see Table I).

Note that this coating design is a suggestion for how to use $SiO_2{:}HfO_2$ calculated based on measurements results of single layers of the different materials. An actual highly reflective multilayer coating is yet to be produced and verified.

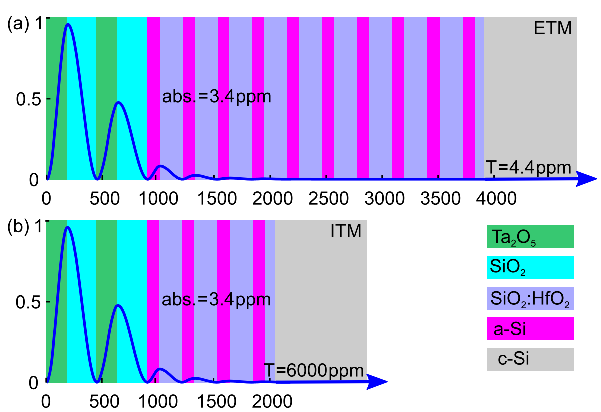

FIG. 4. Design of an ETM and an ITM using a-Si and $SiO_2{:}HfO_2$ capped with two bilayers of SiO_2/Ta_2O_5 to reduce absorption. The layer closest to the substrate is 0.2 QWL thick. All other layers are one QWL thick. The blue line shows the electric field intensity of the laser beam.

Conclusion.—We have shown 30% $SiO_2{:}HfO_2$ to be an excellent low-index material for use in highly reflective mirror coatings together with a-Si. Unlike pure HfO_2, $SiO_2{:}HfO_2$ is stable against crystallization for heat treatment up to 600 °C, which prevents excess scattering—essential for materials to be suitable for gravitational-wave detectors. The mechanical loss of $SiO_2{:}HfO_2$ at a temperature of 10 K is significantly lower than observed for pure SiO_2. After heat treatment at 400 °C, which is the optimum temperature to minimize the optical absorption of a-Si, the mechanical loss of $SiO_2{:}HfO_2$ is more than a factor of 2 below that of SiO_2.

A multimaterial coating made of a-Si and $SiO_2{:}HfO_2$, with two bilayers of SiO_2 and Ta_2O_5 on top, has been demonstrated to fully meet the requirements of ET-LF on CTN_D [54], and on optical absorption for the first time.

There are many other challenges to be overcome to realize the cryogenic Einstein Telescope, but this coating design is an important step towards the detector being able to meet its goal of a factor of 100 improvement in sensitivity over aLIGO at frequencies around 10 Hz.

We are grateful for the financial support provided by STFC under Grants No. ST/L000946/1, No. ST/N005422/1, and ST/N005406/2, by the Royal Society (Grant No. RG110331), and by the University of Glasgow. I. W. M. is supported by a Royal Society Research Fellowship. We are grateful to the International Max Planck Partnership for “Measurement and Observation at the Quantum Limit” for support, and we thank our colleagues in the LSC and Virgo collaborations and within SUPA for their interest in this work. We thank M. Hart for the helpful comments. We would like to thank M. Pitkin for help with low frequency gravitational-wave sources. We would like to thank the referees for support and for bringing order into our noise sources. This paper has LIGO Document number LIGO-P1800241.

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