Demonstration of the Multimaterial Coating Concept to Reduce Thermal Noise in Gravitational-Wave Detectors

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Thermal noise associated with the mechanical loss of current highly reflective mirror coatings is a critical limit to the sensitivity of gravitational-wave detectors. Several alternative coating materials show potential for reducing thermal noise, but cannot be used due to their high optical absorption. Multimaterial coatings have been proposed to enable the use of such materials to reduce thermal noise while minimizing their impact on the total absorption of the mirror coating. Here we present experimental verification of the multimaterial concept, by integrating aSi into a highly reflective SiO_2 and Ta_2O_5 multilayer coating. We show a significant thermal noise improvement and demonstrate consistent optical and mechanical performance. The multimaterial coating survives the heat treatment required to minimize the absorption of the aSi layers, with no adverse effects from the different thermomechanical properties of the three materials.

Introduction.—A number of gravitational-wave signals from binary black-hole mergers [1–5] and from a neutron-star merger [6] have been detected during the first two observing runs of the Advanced LIGO [7] and Advanced Virgo [8] gravitational-wave detectors. At their most sensitive frequencies, these detectors are limited by thermal noise arising from the amorphous highly reflective (HR) multilayer coatings applied to the interferometer mirrors. Several more-sensitive detectors are planned [9,10], aiming to establish black hole populations, potentially uncover deviations from general relativity, clarify the expansion rate of the Universe and investigate the nature of black holes and other exotic sources [9].

All future detectors will require improved optical coatings to enable current coating thermal noise limits to be surpassed. The critical coating properties are the optical absorption, and the mechanical loss to which the thermal noise power spectral density is proportional. The coatings currently used in Advanced LIGO and Advanced Virgo are made from silica (SiO_2) and tantala doped with titania ($TiO_2 : Ta_2O_5$) [7,8,11–14]. Many approaches to develop coatings with reduced thermal noise and low optical absorption at the ppm level are being investigated, such as understanding correlations between atomic structure and material properties [15–19], elevated temperature deposition [20], different dopants and doping concentrations [13,21], nanolayer structures [22,23], crystalline coatings [24], alternative amorphous materials [21,25,26], and optimizing the layer thicknesses [27].

Amorphous silicon (aSi) is one of the most promising options to replace Ta_2O_5 in HR coatings. It can have very low mechanical loss [28] and the high refractive index enables significantly thinner coatings to be made, which further reduces the thermal noise. However, aSi shows too high optical absorption. Recent work has shown that the absorption can be reduced by heat treatment [29], by using a wavelength of 2 μm [30] and by optimizing the deposition procedure [31], but it is still not low enough to meet the requirements of gravitational-wave detectors.

Another option that has been proposed to allow the use of aSi, even with current absorption levels, is a "multimaterial" coating design [32,33]. In such a design, some low-absorption layers are used at the top of the coating stack, to reflect the majority of the incident laser power. Higher absorbing materials are used in the lower parts of the stack where there is little laser power left to be absorbed. This allows the replacement of some relatively high-loss Ta_2O_5 layers with low-loss aSi layers, reducing the total mechanical loss (and thus thermal noise) of the coating stack. Several multimaterial coating designs have been proposed [25,26,31,34], but not experimentally demonstrated.

In practice, multilayer coatings are complex physical systems—which get more complicated the more different materials are involved. In particular, the differing thermal-expansion coefficients of each material make it interesting to investigate whether heat treatment damages the coating. Furthermore, there is evidence that material properties such

as mechanical loss can be altered by stress [35]. The optical absorption might be affected by interface effects. Therefore, an experimental test is required to verify the theoretical prediction of how the various layers interact.

In this Letter, we experimentally prove the multimaterial coating concept by presenting mechanical loss and optical absorption measurements made on a prototype multimaterial coating. This coating is made of 5 bilayers of SiO_2 and Ta_2O_5 followed by 5 more bilayers in which the Ta_2O_5 is replaced by aSi; see Fig. 1. Throughout this Letter we refer to the 5 double layers of SiO_2/Ta_2O_5 as upper stack, to the 5 double layers of SiO_2/aSi as lower stack and to all 10 double layers as full stack.

By comparing to our upper stack, we show that the incorporation of aSi layers reduces thermal noise of the full multimaterial stack by a factor of 2 compared to a pure SiO_2/Ta_2O_5 coating with approximately the same reflectivity (deposited using the same technique). Because of the reflectivity of the upper stack, the absorption in the full multimaterial coating is more than a factor 20 below that of a pure SiO_2/aSi coating. The absorption behaves similarly with heat treatment to the absorption observed in an aSi/SiO_2 bilayer [29]. Heat treatment up to 600 °C, which is above the temperature at which the absorption of aSi minimizes, is possible without causing damage (e.g., cracks) to the coating.

Coating transmission.—The coatings studied here were deposited using an ion plating technique [36] that can produce aSi with low optical absorption at 2 μm and low mechanical loss [30]. Disks of diameter 76.2 mm and thickness 2.54 mm were used for the mechanical loss measurements [37]. For the absorption measurement, samples of diameter 25.4 and thickness 6.35 mm were used. The majority of absorption samples were made of Corning 7980 silica. Corning 7980 has significant absorption at 2 μm, making it an unsuitable substrate for absorption measurements of our low-absorption upper stack. Therefore a few Corning 7979 samples, which have lower absorption at 2 μm due to lower OH content, were used.

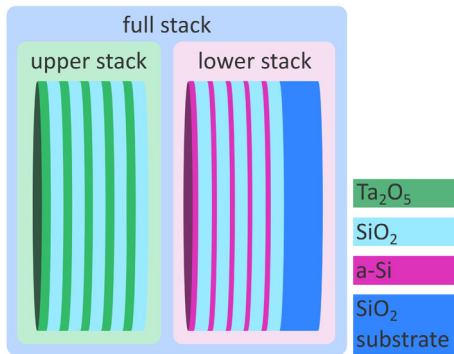The lower stack was deposited onto a large set of samples. After deposition, some samples were removed from the coating chamber (providing the lower stack sample set), and replaced with new, uncoated samples. The upper stack was then deposited. This resulted in a set of samples coated with each of the three stacks, and ensured that the two components of the full-stack coating were identical to the separate upper and lower stacks.

The full stack was designed to provide a transmission of < 10 ppm, similar to end test masses used in gravitational-wave detectors. Figure 2 shows measured spectra of the three coatings in an as-deposited state. While the transmission T was measured, the graph shows $1 - T$, which is equivalent to the sum of reflectivity R and absorption A, with $R + T + A = 1$. On the scale used in this plot, A is negligibly small.

The pink line in Fig. 2 shows $1 - T$ for the lower stack. Because of the high refractive index of aSi, this coating shows a broad, highly reflective plateau. The green line shows $1 - T$ for the upper stack. Because of the lower refractive index of Ta_2O_5, the reflectivity of this coating is significantly lower than that of the lower stack, with a narrower plateau. The blue line shows $1 - T$ of the full stack.

Optical absorption.—To reduce their optical absorption, HR coatings often get heat treated following deposition [38]. Previous work indicates that the absorption of aSi is particularly strongly dependent on heat treatment, and can be reduced by a factor of up to 50 by heating to a temperature between 400 °C and 500 °C [29–31]. The exact temperature at which the minimum absorption occurs, and the improvement factor, vary slightly depending on the coating deposition procedure used.

To investigate the effect of heat treatment on the multimaterial coating, we heat treated samples coated with the full stack and the lower stack at 100 °C, 200 °C, 300 °C, 400 °C, 500 °C, and 600 °C. For consistency checks, the absorption of the upper stack was also measured for the as-deposited coating and after heat treatment at 500 °C [40]. Heat treatment was carried out for 3 h in air. Coatings can
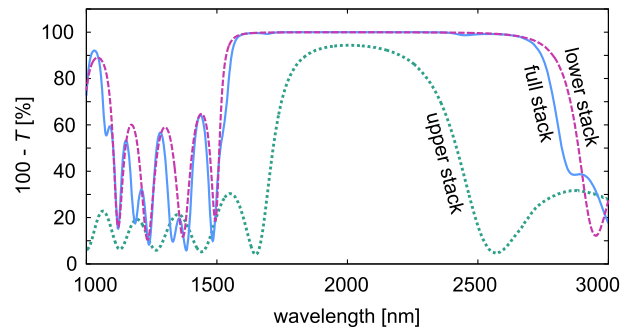

FIG. 1. Schematic of the multimaterial coating: The full stack is composed of the upper stack (SiO_2 and Ta_2O_5) and the lower stack (aSi and SiO_2).

FIG. 2. Spectra of the three coatings: the y axis shows 100% minus the measured transmission T. This is equivalent to the sum of the reflection R and the absorption A—which can be assumed to be small on this scale and in this wavelength range.

sometimes exhibit damage such as cracking or delamination, particularly following heat treatment. Microscope images of all samples were taken, and no evidence for either cracking or delamination was observed.

The optical absorption of the three stacks was measured at 2 μm using photothermal common-path interferometry [41], which is an optical technique for measuring the thermal effect of absorbed laser power.

Figure 3 shows the measured absorption as a function of heat-treatment temperature for the lower stack (pink squares), the upper stack (green diamonds), and the full stack (blue circles). None of the coating stacks shows a significant change in absorption at heat-treatment temperatures of $\leq 200\,°C$. We believe that this is because the substrates reach a temperature of $\approx 200\,°C$ during deposition, and therefore no changes occur with further heat treatment at or below this temperature. For heat treatment above $200\,°C$, the absorption of the lower stack reduces by a factor of ≈ 7 to a minimum value of 181 ppm at $500\,°C$. The upper stack reduces from 4.8 to 3.0 ppm. The full stack also reduces in absorption, by a factor of ≈ 5 to a minimum value of 8.1 ppm at $500\,°C$, which is a factor of 22 lower than the absorption of the lower stack.

To allow comparison of our measurements with other coatings, it is necessary to calculate the extinction coefficient k of the coatings. k, to which the absorption is proportional, is the imaginary part of the refractive index. For aSi, k was estimated from the absorption of the lower stack under the assumption that the absorption of SiO_2 is negligible compared to aSi. Similarly, k of Ta_2O_5 was estimated from the absorption of the upper stack. The light field in the full stack was simulated using these k values to predict the absorption of the full stack from the two partial stacks. This prediction is shown by the red circles in Fig. 3 for the coatings as-deposited and heat treated at $500\,°C$ ($k \approx 3.4 \times 10^{-4}$ for aSi and $k \approx 1.2 \times 10^{-6}$ for Ta_2O_5), and agrees closely with the measured absorption of the full stack.

Mechanical loss.—Fused silica disks coated with each of the three stacks were mounted on a nodal support [42], in which the disk is balanced on a silicon spherical cap of radius of curvature 60.44 mm. The mechanical loss of the three coating stacks was measured using a ring-down technique, in which the free-amplitude decay of vibrational modes of the disks is measured. For vibrational modes with a node at the center of the disk, this is a highly effective method of minimizing external frictional damping and ensuring that only losses due to internal friction in the disk are measured. The mechanical loss of approximately eight modes of each disk, between ≈ 2.8 and ≈ 30 kHz, was measured before and after deposition of the coatings. To obtain the coating loss, the difference in coated and uncoated loss was scaled by the ratio of the elastic energy stored in the substrate to that stored in the coating [43]. This ratio was calculated using finite element analysis (COMSOL). Figure 4 shows a photograph of a disk coated with the full stack in the setup used for loss measurements and of a COMSOL model of an excited mode of the disk.

The loss was found to have some variation with frequency, which is likely to partially originate from a different split into bulk and shear motion dependent on the mode shape [44]. Figure 5 shows the average coating loss, calculated over all measured modes, for each stack as a function of heat treatment—see Table I for the results of the coating as deposited and after heat treatment at $500\,°C$. As expected, the lower stack has the lowest loss, due to the low loss of the SiO_2 and aSi layers. The loss of the upper stack is significantly higher, due to the high loss of the Ta_2O_5 layers.

The loss of all three stacks decreased with heat treatment—by 17% for the upper stack, by 36% for the lower stack, and by 26% for the full stack. This trend is as predicted from previous measurements of the individual materials [20,30,49]. Also shown in Fig. 5 is the loss of the full stack predicted from the measurements of the upper and lower stacks (red circles). The prediction shows excellent agreement with the measurements (blue dots). The average loss of the full stack, at the optimum heat-treatment temperature, is
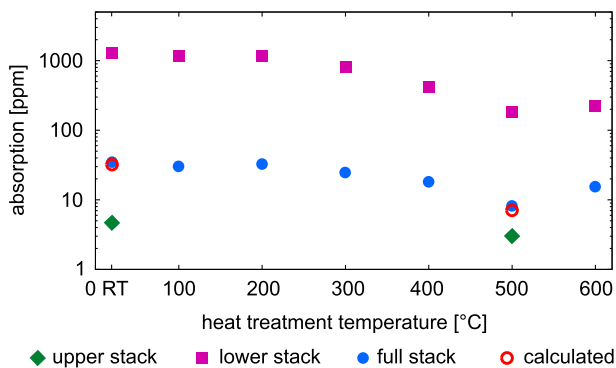

FIG. 3. Measured absorption of the three coatings as a function of heat-treatment temperature. Also shown is the predicted absorption of the full stack, based on the measurements of the upper and lower stacks. (Note that the samples reach a temperature of $\approx 200\,°C$ during deposition.)
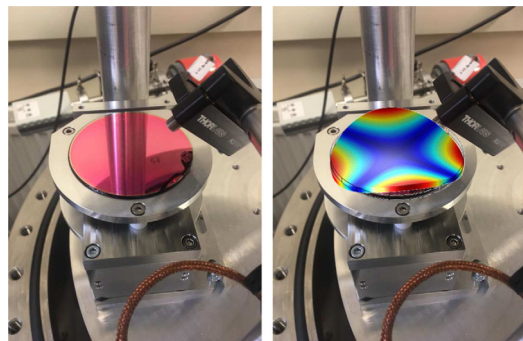

FIG. 4. Photo of the full stack multimaterial coating in the setup for mechanical loss measurements (left) and a COMSOL simulation of the disk with an excited butterfly mode at 2.7 kHz (right).
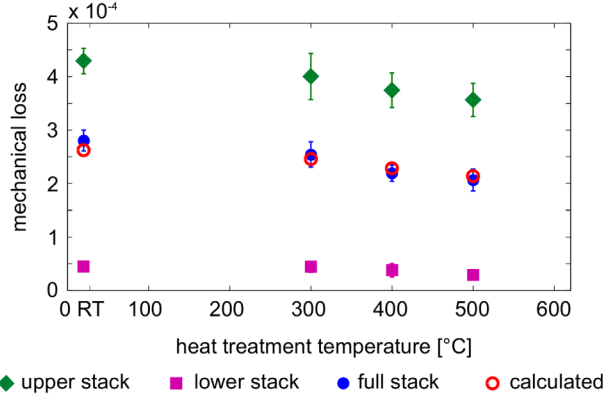
FIG. 5. Measured mechanical loss of the three stacks as a function of heat-treatment temperature. Also shown is the predicted loss of the full stack calculated from the measurements of the upper and lower stacks. (Note that the samples reach a temperature of $\geq 200\,°C$ during deposition.)

$\approx 42\%$ lower than the loss of the upper stack, demonstrating the value of including aSi in the coating design [50].

Coating thermal noise.—In addition to loss, the thickness of a mirror coating is a crucial parameter in determining the thermal noise, and the high index of aSi brings significant benefits in enabling a thinner coating to achieve the same reflectivity. Thermal noise also depends on the Young's moduli of the coating materials. In this section, we present thermal noise calculations to compare the overall performance of our coatings.

Coating thermal noise amplitude spectral density is given by [33]

$$ x(f) = \sqrt{\frac{2k_B T}{\pi^2 f}\frac{1}{w^2}\frac{1 - \sigma_{\text{sub}} - 2\sigma_{\text{sub}}^2}{Y_{\text{sub}}} \sum_j b_j d_j \phi_j}, \quad (1) $$

where k_B is the Boltzmann constant, T the mirror temperature, f the frequency, and w the radius of the interferometer laser beam on the coating. Y_{sub} and σ_{sub} are the Young's modulus and the Poisson ratio of the substrate. d is the

coating thickness and ϕ the coating mechanical loss. The index j refers to the material parameters of the jth layer in the coating (starting from the outermost layer). For the calculations, we assume here that the mechanical losses associated with bulk motion and shear motion [44] are approximately equal ($\phi_{\text{bulk}} \approx \phi_{\text{shear}} \approx \phi$). b_j (The first factor in square brackets is a correction [51] to the expression for b_j given by Yam, with which the authors of the Yam paper agree) is a weighting factor described by

$$ b_j = \left[\frac{(1 - 2\sigma_j)(1 + \sigma_j)}{(1 - 2\sigma_s)(1 + \sigma_s)}\right]\frac{1}{1 - \sigma_j} $$

$$ \times \left[\left(1 - n_j\frac{\partial \theta_c}{\partial \theta_j}\right)^2\frac{Y_s}{Y_j} + \frac{(1 - \sigma_s - 2\sigma_s^2)^2}{(1 + \sigma_j)^2(1 - 2\sigma_j)}\frac{Y_j}{Y_s}\right], \quad (2) $$

where σ_j and σ_s are the Poisson's ratios of each coating layer and the substrate. n_j is the refractive index of the jth layer and $\partial \theta_{\text{coat}}/\partial \theta_j$ describes the sensitivity of the total coating phase θ_{coat} to fluctuations in the round-trip phase θ_j in each layer.

$x(f)$ was calculated for 19 bilayers of SiO_2/Ta_2O_5 which provide a similar reflectivity to our full multimaterial stack ($\approx 99.999\%$). We used the average Young's modulus of the two materials and the loss measured on our upper stack after heat treatment at $500\,°C$ (see Table I).

Layers contribute more to coating thermal noise the lower in the stack they are located, as discussed by Ref. [44] in the derivation of Eq. (2). This is relevant here due to the lower loss of the lower stack. The coating thermal noise of the multimaterial coating reduces by $(49.4 \pm 0.5)\%$ compared to the pure SiO_2/Ta_2O_5 coating with similar reflectivity.

Summary.—We have measured the optical absorption and the mechanical loss of a multimaterial coating consisting of an upper stack of SiO_2/Ta_2O_5 and a lower stack of SiO_2/aSi. We showed that the results for the partial stacks and the full stack are self-consistent. Furthermore, we observed no damage evidence of cracking or delamination

TABLE I. Measured losses for our three stacks and material parameters used for calculating coating thermal noise. For the individual materials, the thickness t of one quarter-wave layer is given. The effective Young's modulus Y and Poisson ratio σ of multilayer coatings was obtained using a thickness-weighted average of the values for each component layer [45].

	Y (GPa)	d (nm)	n at 2000 nm[a]	σ	$\phi \times 10^{-4}$ As deposition	$\phi \times 10^{-4}$ 500 °C
SiO_2	72 [46]	347	1.44	0.17 [47]		
Ta_2O_5	140 [46]	236	2.12	0.23 [9]		
aSi	147 [48]	134	3.73	0.23		
Upper stack	99.5	5 × (347 + 236) = 2915		0.2	(4.29 ± 0.24)	(3.56 ± 0.31)
Lower stack	92.9	5 × (347 + 134) = 2405		0.2	(0.44 ± 0.06)	(0.28 ± 0.06)
Full stack	96.3	2915 + 2405 = 5320		0.2	(2.80 ± 0.20)	(2.06 ± 0.21)

[a]measured for aSi (which dominates the reflectivity) single layers produced by the same vendor via fitting of transmission spectra: n changes by $< 2\%$ with heat treatment and $n \times t$ by $< 1\%$.

due to the differing thermomechanical properties of the different coating materials. Our coating provides almost a factor of 2 less thermal noise compared to a full SiO_2/Ta_2O_5 coating deposited using the same process, while successfully suppressing the optical absorption contribution of the SiO_2/aSi layers by more than a factor of 20.

This coating design was optimized to demonstrate the principle of multimaterial coatings and is unlikely to be suitable for use in a certain gravitational-wave detector as it is, as it was not designed and optimized for this purpose. This experimental verification is a significant step towards the realization of proposed multimaterial coating designs for the next generations of gravitational-wave detectors [25,26,31,34] to enable the required thermal noise reduction.

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