Low Mechanical Loss math xmlns="http://www.w3.org/1998/Math/MathML" display="inline">mrow>mrow>msub>mrow>mi>TiO/mi>/mrow>mrow>mn>2/mn>/mrow>/msub>/mrow>mo>:/mo>mrow>msub>mrow>mi>GeO/mi>/mrow>mrow>mn>2/mn>/mrow>/msub>/mrow>/mrow>/math> Coatings for Reduced Thermal Noise in Gravitational Wave Interferometers

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Low mechanical loss TiO_2:GeO_2 coatings for reduced thermal noise in Gravitational Wave Interferometers

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The sensitivity of current and planned gravitational wave interferometric detectors is limited, in the most critical frequency region around 100 Hz, by a combination of quantum noise and thermal noise. The latter is dominated by Brownian noise: thermal motion originating from the elastic energy dissipation in the dielectric coatings used in the interferometer mirrors. The energy dissipation is a material property characterized by the mechanical loss angle. We have identified mixtures of titanium dioxide (TiO_2) and germanium dioxide (GeO_2) that show internal dissipations at a level of 1×10^{-4}, low enough to provide almost a factor of two improvement on the level of Brownian noise with respect to the state-of-the-art materials. We show that by using a mixture of 44% TiO_2 and 56% GeO_2 in the high refractive index layers of the interferometer mirrors, it would be possible to achieve a thermal noise level in line with the design requirements. These results are a crucial step forward to produce the mirrors needed to meet the thermal noise requirements for the planned upgrades of the Advanced LIGO and Virgo detectors.

Gravitational wave (GW) detectors are highly sensitive instruments that measure the very small distance changes produced by signals of astrophysical origin [1, 2]. The current generation of GW detectors are km-scale laser interferometers [3–6] with several hundreds of kW of circulating power in the Fabry-Perot arm cavities. The test-mass mirrors are made of high-purity fused silica substrates, coated with high-reflectivity multilayer dielectric thin-film stacks [7], composed of multiple pairs of high and low refractive index metal oxide layers, making a Bragg reflector structure.

The sensitivity of the current detectors [8, 9] is limited by a combination of laser quantum noise [10] and displacement noise generated by the Brownian motion of the coatings [11, 12]. Therefore, to increase the astrophysical reach of future detectors, it is crucial to reduce coating Brownian noise. This in turn requires reducing the elastic energy dissipation in the thin film materials composing the coatings [12, 13]. The power spectral density of Brownian noise at a frequency f is a complex function of the properties of the materials used in the coatings [14, 15]. An approximate expression, assuming equal bulk and shear loss angles, is given by (see [15] and supplemental material [16]):

$$S_\mathrm{B}(f) = \frac{2k_BTd}{\pi^2 w^2 f}\left[\left\langle \frac{Y}{1-\nu^2}\phi\right\rangle \frac{(1+\nu_S)^2(1-2\nu_S)^2}{Y_S^2} + \left\langle \frac{(1+\nu)(1-2\nu)}{(1-\nu)Y}\phi\right\rangle\right] \quad (1)$$

where k_B is the Boltzmann's constant, T is the ambient temperature, w is the radius of the laser beam probing the mirror motion, d is the total thickness of the coating, Y_S and ν_S are the Young's modulus and Poisson ratio of the substrate. The angular bracket expression $\langle x\rangle$ indicates the *effective medium* average [15, 17] of the material property x through the stack, weighted by the physical thickness of the layers. The relevant properties of the coating materials are the Young's moduli Y, the Poisson ratios ν and the loss angles $\phi = \mathrm{Im}(Y)/\mathrm{Re}(Y)$.

The coatings used in the current Advanced LIGO mirrors are composed of alternating layers of amorphous SiO_2 of low refractive index $n_{\mathrm{SiO_2}} = 1.45$ at 1064 nm, and TiO_2:Ta_2O_5 of high refractive index $n_{\mathrm{TiO_2:Ta_2O_5}} = 2.10$ at 1064 nm [18, 19]. The TiO_2:Ta_2O_5 layers have a loss angle much larger than the SiO_2 layers ($3-4\times10^{-4}$ [20, 21] compared to $\sim 2\times10^{-5}$ [18]) and therefore they dominate in the contribution to the coating Brownian noise.

The goal for the next upgrade to the LIGO detectors, called Advanced LIGO+ [22, 23] is a reduction of the coating noise by about a factor of two, with a target Brownian noise of $S_\mathrm{B}^{1/2} = 6.6\times10^{-21}\mathrm{m}/\sqrt{\mathrm{Hz}}$ at a frequency of 100 Hz. The SiO_2 layers can already be produced with low enough mechanical loss angle [18], so the main focus of the current research is on improving the high refractive index material. Several different approaches have been investigated, including deposition at elevated substrate temperatures [24, 25] and with assist ion bombardment [26, 27], doping and nanolayering of Ta_2O_5 [28–31], and the use of nitrides [32, 33]. Here we report results on amorphous oxide coatings based on mixtures of GeO_2 and TiO_2.

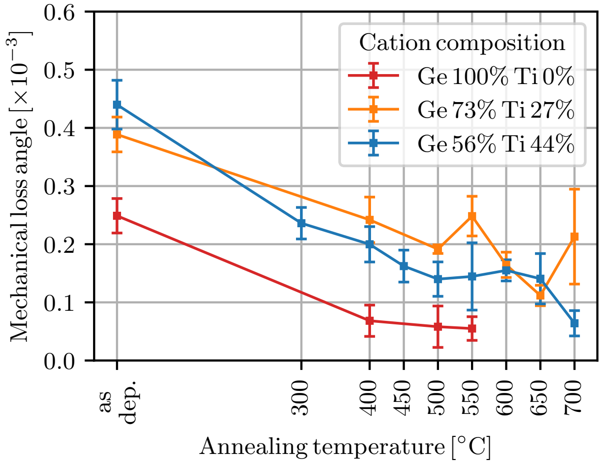

FIG. 1. Measured loss angle of TiO_2:GeO_2, as deposited and after 10-hours-long annealing in air, at increasing temperatures. Different color lines correspond to the cation composition listed in the legend. Only one sample for each concentration is shown here for simplicity. Other samples showed equal values within the error bars.

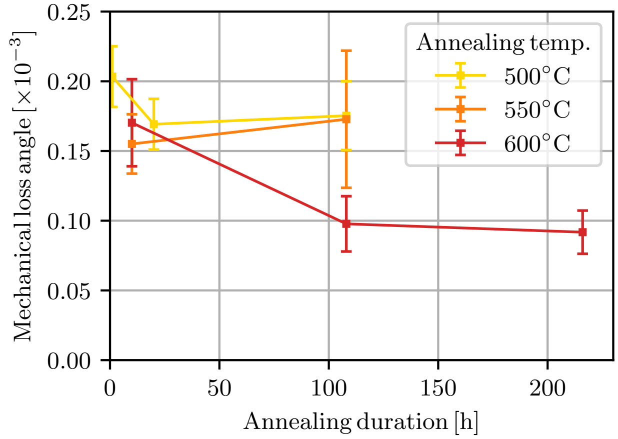

FIG. 2. Effect of the annealing duration on the measured loss angle for the 44% TiO_2:GeO_2 film.

The initial motivation to investigate coatings based on GeO_2 was the discovery of a correlation between the room-temperature mechanical loss angle and the fraction of edge-sharing versus corner-sharing polyhedra in the medium-range order, as reported in [34] for ZrO_2:Ta_2O_5. SiO_2 also has a prevalence of corner-sharing, and is the amorphous material that exhibits the lowest known room-temperature loss angle in the acoustic frequency range [18, 35, 36]. Additionally, the mechanical loss angle of GeO_2 at low temperatures [37] ($\lesssim 100$ K) exhibits a peak similar to the one found in SiO_2 [38, 39]. In recent experiments on GeO_2 [40], we confirmed that the atomic packing can be altered to improve medium range order by annealing and high temperature deposition. Similar correlations were found for different oxides by other groups [41–45].

From the optical perspective, however, GeO_2 has a refractive index $n = 1.60$ at 1064 nm that makes it unsuitable for use in a high reflector design when combined with SiO_2, as 138 layers would be needed to achieve the required high reflectivity for the test-mass mirrors. The increase in the total thickness of a GeO_2/SiO_2 reflector would balance out the reduced mechanical loss, with no net improvement in the coating Brownian noise. To increase the refractive index at the laser wavelength of 1064 nm, GeO_2 was co-deposited with TiO_2 with different cation concentrations.

Thin films of TiO_2:GeO_2 with Ti cation concentration of 0%, 27%, and 44%, were deposited by ion beam sputtering using a biased target deposition system [46], that allowed convenient tuning of the mixture composition by adjusting the length of the pulses biasing the metallic Ti and Ge targets.

The cation concentration, oxygen stoichiometry and atomic areal density of the films were determined by Rutherford backscattering spectrometry (RBS) [47]. The thickness and refractive index were obtained from spectroscopic ellipsometry. The mass density was computed from the RBS and ellipsometry measurements. The absorption loss at the wavelength of 1064 nm was assessed from photo-thermal common-path interferometry [48]. The thin films were annealed in air, as annealing has been shown to reduce absorption loss and room-temperature mechanical loss angle in amorphous oxides [49, 50]. Grazing incidence x-ray diffraction shows all mixture films are amorphous upon annealing at 600°C for 10 and 108 hours, and show signs of crystallization when annealed at higher temperatures. The pure GeO_2 film remained amorphous up to 550°C.

For the Ti cation concentration of 44%, the refractive index at 1064 nm is $n_{TiO_2:GeO_2} = 1.88$. The absorption loss normalized to a quarter-wavelength (QWL) thick single layer (141 nm) is 2.3 ± 0.1 ppm after annealing at 600°C. The absorption loss of pure GeO_2 after annealing at 500°C is below 1 ppm at $\lambda = 1064$ nm, showing the potential for improved absorption in the mixture. The deposition parameters are being optimized to achieve even lower optical absorption in TiO_2:GeO_2, to meet the Advanced LIGO+ requirements of less than 1 ppm [23] for a full mirror coating. More details on the structural and optical characterizations are available in the supplemental material [16].

The thin films were also deposited with the same procedure on 75-mm-diameter, 1-mm-thick silica disks, to measure the material's elastic properties. The disk acts as a resonator: about 20 modes between 1 kHz and 30 kHz can be measured in a Gentle Nodal Suspension [51, 52] to obtain their precise frequency and decay time. After the thin film is deposited on the substrate, the resonant frequencies are shifted by amounts depending on the film properties, allowing an estimation of the Young's modulus and Poisson ratio [18, 53]. The decay times of the modes of the coated substrates are significantly shorter than for the bare substrate, due to the elastic energy dissipation in the film. Using the measured elastic properties of the film material, one can compute the fraction of elastic energy in the film for each resonant mode and use it to extract the loss angle ϕ of the thin film material [54].

For a homogeneous amorphous material, the relation between stress and strain in the elastic regime can be described in terms of two elastic moduli, for example bulk K and shear μ moduli [55]. Similarly, the internal energy dissipation in the material should be described in terms of two loss angles $\phi_K = \mathrm{Im}(K)/\mathrm{Re}(K)$ and $\phi_\mu = \mathrm{Im}(\mu)/\mathrm{Re}(\mu)$. There is no physical reason to assume the two loss angles to be equal, and we shall show in the following that they are indeed significantly different for TiO_2:GeO_2. The layered structure of the stack implies that a description in terms of an equivalent isotropic material is not accurate, since the bulk and shear energy distribution in the layers is different in the case of the ring-down measurements and in the Brownian noise case. While the expression in equation 1 assumes equal loss angles, a more precise expression, including the distinction between bulk and shear properties for all layers, is described in the supplemental material [16], and is needed to correctly account for the multilayer structure and the different materials.

However, in the initial exploration of the effect of composition and annealing schedule, we relied on the commonly used description with frequency independent equal bulk and shear loss angles [18, 56, 57]. The more detailed analysis of the best candidate material, described later, supports the use of this simplification for survey purposes, since our measurements are more sensitive to the shear than the bulk loss angle, and the former is found to be almost frequency independent. With this approach, figure 1 shows the measured loss angle for pure GeO_2 and the two concentrations of TiO_2 and GeO_2 studied in detail here. The most promising results are from a mixture of 44% TiO_2 and 56% GeO_2. The mechanical loss of amorphous oxides typically decreases with increasing annealing temperature and time. We observed rapid crystallization at 700°C, and therefore explored the effect of annealing duration on the loss angle. We tested heat treatments of 1, 10, 20, 108 and 216 hours in total, for temperatures of 500, 550 and 600°C. Figure 2 shows the effect of annealing time on the loss angle of the 44% TiO_2:GeO_2 mixture. It was found that extended annealing at lower temperatures produces little improvement. Instead, after annealing at 600°C for 108 hours, the loss angle is reduced to $(0.96 \pm 0.18) \times 10^{-4}$, and the film is still amorphous. Among those tested in our work, this TiO_2:GeO_2 mixture is the most promising high-index material for low Brownian noise Advanced LIGO+ mirrors, though further characterization of mixtures with other Ti/Ge ratios in this range is planned to find the optimum.

SiO_2 property	**Value**
Refr. index at 1064 nm	1.45 ± 0.01
Young's modulus	73.2 ± 0.6 GPa
Poisson ratio	0.11 ± 0.07
Loss angle	$\phi_K = \phi_\mu = \left(2.6^{+0.5}_{-0.6}\right) \times 10^{-5}$

TiO_2:GeO_2 property	**Value**
Cation conc. Ti/(Ti+Ge)	44.6 ± 0.3 %
Refr. index at 1064 nm	1.88 ± 0.01
Optical abs. for a QWL	2.3 ± 0.1 ppm
Density	3690 ± 100 kg/m^3
Young's modulus	91.5 ± 1.8 GPa
Poisson ratio	0.25 ± 0.07
Bulk Loss angle	$a_K = \left(22.0^{+10.6}_{-12.5}\right) \times 10^{-5}$ $m_K = 1.04^{+0.40}_{-0.36}$
Shear Loss angle	$a_\mu = \left(8.4^{+2.9}_{-4.0}\right) \times 10^{-5}$ $m_\mu = -0.06^{+0.15}_{-0.30}$

TABLE I. Measured parameters for TiO_2:GeO_2 and SiO_2, after annealing at 600°C for 108 hours. The loss angle model for TiO_2:GeO_2 is $\phi(f) = a \cdot (f/10\,\mathrm{kHz})^m$. Uncertainties describe the 90% confidence intervals.

As a first step toward the production of a full high-reflectivity coating, and to better characterize this new material, we deposited single layers of SiO_2 and TiO_2:GeO_2, as well as a stack of 5 QWL layers of TiO_2:GeO_2 alternated with 5 layers of SiO_2, and 20 layers of TiO_2:GeO_2 alternated with 20 layers of SiO_2. The depositions were performed using a commercial Spector Ion Beam Sputtering system that can produce films with better optical quality [26] than the biased target system used for the initial parameter exploration. At the laser wavelength of 1064 nm, the transmission of the 40-layer structure was 190 ppm and the optical absorption was measured to be 3.1 ppm after annealing. We also measured the Young's modulus and loss angle of the stacks. However, since the multilayers structure is not isotropic, a description in terms of Y and ν is only approximate. Nevertheless, the two stacks were found to have the same Young's modulus, 78.0±1.3 GPa, and the same loss angle, $(5.5\pm0.7)\times10^{-5}$ after annealing at 600°C for 108 h. This is an indication that there is no evidence of any systematic error in the measurements due to the thickness of the coatings. At an approximation level consistent with assuming equal bulk and shear loss angles, one can compute the expected value for the stack by averaging the single material values as $\bar{\phi} = \langle Y\phi \rangle / \langle Y \rangle = (6.4 \pm 1.7) \times 10^{-5}$. Therefore there is no indication of excess loss due to interfaces [58].

A correct description of the Brownian noise in a multilayer stack must take into account the bulk and shear moduli and loss angles of the individual materials. The resonant modes of the coated disk store different fractions

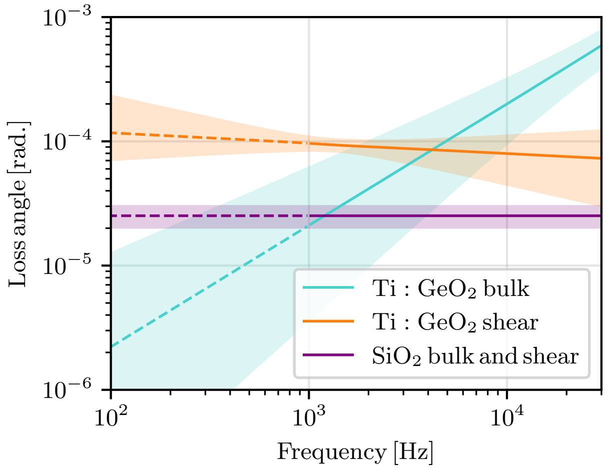

FIG. 3. Estimated bulk and shear loss angles as a function of frequency. The solid lines indicate the range of frequencies where the loss angles were measured, while the dashed lines are extrapolations to the lower frequency range. The shaded regions show the 90% confidence intervals of the estimates.

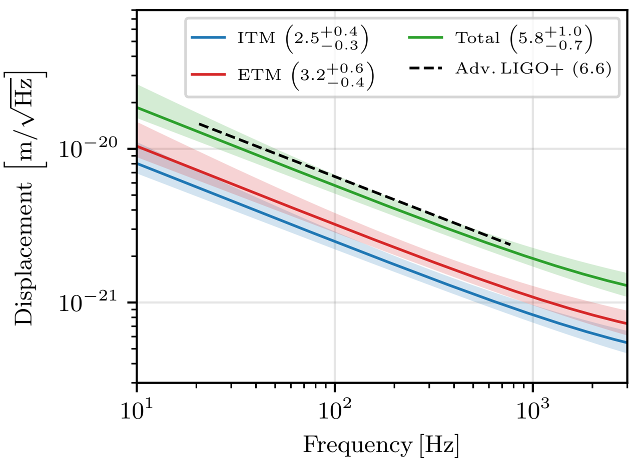

FIG. 4. Estimated Brownian noise for the Advanced LIGO+ interferometer. The red and blue traces show the contribution of a single ITM and ETM, while the green trace shows the total for all four test masses. The numbers in the legend give the Brownian noise level at 100 Hz, in units of $10^{-21}\mathrm{m}/\sqrt{\mathrm{Hz}}$. The shaded regions correspond to the 90% confidence intervals of the estimates. The dashed black line shows the design target for Advanced LIGO+.

of bulk and shear energy in the film, and therefore it is possible to extract the bulk and shear loss angles from the measurements [53, 59]. We model a single isotropic layer with known thickness and density as measured by ellipsometry and RBS, and with Young's modulus, Poisson ratio and bulk and shear loss angles as free parameters. For each sample, the measurement data set consists of the frequency shifts due to the coating, and the reduction in the decay time, due to the energy dissipation in the coating, for each of the measurable modes. We used a Markov chain Monte Carlo Bayesian Analysis [53, 60, 61] to find the probability distribution of the model parameters given the data. We considered either different bulk and shear loss angles or equal loss angles, and three possible frequency dependencies: constant, linear or power law, for a total of six different loss models. The Bayesian analysis allows us to compute the relative likelihood of each model given the data. The best model for the TiO_2:GeO_2 film is a power law with different bulk and shear loss angles, while for the SiO_2 film it is a constant single loss angle, as shown in figure 3. It is worth noting that the bulk loss angle for the TiO_2:GeO_2 film shows a rather steep frequency dependence. The second most likely model for this material is the one with a linear frequency dependence. The bulk loss angle does not show a frequency dependency as steep as in the power law case, but it is still predicted to be significantly smaller than the shear loss angle at low frequency. The measured value of the loss angle for SiO_2 is compatible with values reported in the literature [18]. Table I summarizes all the measured material properties. More details on the analysis and the results are in the supplemental material [16].

The transmission requirements for the Advanced LIGO+ test masses are similar to those for Advanced LIGO: the input mirror test masses (ITM) should have a transmission of 1.4% and the end test masses (ETM) of 5 ppm [3]. Given the measured refractive indexes, the ITM stack is composed of 11 layers of 106 nm of TiO_2:GeO_2 alternated with 11 layers of 228 nm of SiO_2, while the ETM stack is composed of 26 layers of 123 nm of TiO_2:GeO_2 and 26 layers of 207 nm of SiO_2. Both structures are capped with a half-wavelength-thick SiO_2 layer.

The Brownian noise for such mirrors can be computed using the effective medium approach described in the supplemental material [16], which has been checked to provide results within a few percent of other published formulas [14, 62]. The results are shown in figure 4. The noise is compliant with the design requirement for Advanced LIGO+, reaching $\left(5.8^{+1.0}_{-0.7}\right)\times 10^{-21}$ $\mathrm{m}/\sqrt{\mathrm{Hz}}$ at 100 Hz. It is worth noting that this result does not depend strongly on the steep frequency dependency predicted for the bulk loss angle of TiO_2:GeO_2. If we use the second most probable model, with a less steep frequency dependency, we obtain $\left(6.2^{+1.5}_{-0.8}\right)\times 10^{-21}$ $\mathrm{m}/\sqrt{\mathrm{Hz}}$ at 100 Hz, still compatible with the Advanced LIGO+ design requirement. Preparation of samples on disks with lower resonant frequencies to better constrain these estimates is underway.

In summary, we demonstrated that a mixture of 44% TiO_2 and 56% GeO_2 offers excellent optical quality and low mechanical loss angle, making it a promising material to be used as high-index layer in the test mass coatings of the Advanced LIGO+ interferometric GW detectors. We analyzed the internal energy dissipation of this novel material in terms of bulk and shear loss angles, and used the results to design multilayer high reflectivity stacks for the Advanced LIGO+ mirrors. The Brownian noise achievable with TiO_2:GeO_2 / SiO_2 based mirrors reaches a level compliant with the Advanced LIGO+ design requirements.

Studies are on-going to further improve mechanical and optical absorption losses by changing deposition parameters, mixture and annealing schedule, and to characterize the scattering properties of the multilayer stacks. We are also planning to directly measure the Brownian noise of optimized high reflection mirrors designed for Advanced LIGO+ [20], to confirm the noise prediction.

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