An Overview of Research into Low Internal Friction Optical Coatings by the Gravitational Wave Detection Community

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The direct detection of gravitational waves by ground-based interferometric gravitational wave detectors in recent years has opened a new window of the universe, allowing the astrophysical observations of previously unexplored phenomena, such as the collisions of black holes and neutron stars. However, small thermodynamic fluctuations of the density of the thin films that compose the mirrors used within the gravitational wave detectors, such as the LIGO and Virgo detectors, give rise to noise which limits these instruments at their most sensitive frequencies. This "Brownian Thermal Noise" can be related to the inherent internal friction of the mirror materials through the fluctuation-dissipation theorem. Therefore, the improved sensitivity of gravitational wave detectors depends, to some extent, upon the development of optical thin films with low internal friction. The past two decades have therefore seen the growth of internal friction experiments undertaken within the gravitational wave detection community. This article attempts to summarize the results of these investigations and to highlight current research directions in order to foster a stronger dialogue with the larger internal friction and mechanical spectroscopy community.

Keywords: LIGO, Internal Friction, Optical Films.

1. The Intersection of Internal Friction and Gravitation Wave Detection

As of the writing of this manuscript, the Advanced LIGO¹ detectors have announced the detection of gravitational wave (GW) signals originating from the inspiral, merger, and ringdown of five binary black hole systems²⁻⁶ and one binary neutron star system⁷. These detections mark the beginning of a new form of astronomy wherein GWs, as opposed to light, provide new information about the universe. While the methods of GW production and propagation lie outside the scope of this manuscript - an excellent description is provided by Ju et al.⁸ - it is instrumental to note that the detected quantity, the GW strain *h*, was in all three cases on the order of 10^{-21} .

Modern interferometric GW detectors, like the Advanced LIGO¹, Advanced Virgo⁹, GEO600¹⁰, and KAGRA¹¹ detectors, use highly sensitive interferometer configurations in order to detect the minute changes in distance between an interferometer's test mass mirrors caused by a passing GW. These distance changes, ΔL , taken over the original test mass separation, L, provide the amplitude of the GW strain: the aforementioned h. Given that the initial test mass separations in these detectors are of the order of 10³ m in all Earth-based GW detectors (4 km in the case of the Advanced

LIGO detectors), it is easy to see that the detectors are able to detect length variations as small as $\Delta L \sim 10^{-18}$ m! With measurements of this scale come noise sources not generally expected in everyday laboratory experiments. A number of the limiting noise sources are shown in Figure 1.

The noise source most relevant to this review is that of coating Brownian thermal noise (CBTN). CBTN is caused by thermally driven random density fluctuations in the coatings used to make the interferometer mirrors. These fluctuations can be related to the internal friction of the coating materials through the fluctuation-dissipation theorem of Callan and Welton¹². The importance of this noise source to the field of GW detection was first elucidated by Levin¹³, and expanded upon by others¹⁴⁻¹⁶. A simplified version of the equation describing the power spectral density of the CBTN for a single mirror, $S_{CBTN}(f)$, where the elastic properties of the mirror coating and substrate are assumed to be equal, and the Poisson ratio assumed to be zero, can be written as:

$$S_{CBTN}(f) \approx \frac{k_B T}{\pi^2 f} \frac{1}{E} \frac{d}{w^2} \phi_{coat}, \qquad (1)$$

where k_{B} is the Boltzmann constant, *T* is the temperature of the mirror, *d* is the thickness of the mirror coating, *f* is the frequency of interest, *E* is the Young's modulus, *w* is the Gaussian beam radius of the laser spot reflecting from the mirror, and ϕ_{coat} is the internal friction of the mirror coating.



Figure 1. Design sensitivity of the Advanced LIGO gravitational wave detectors with the limiting noise sources plotted separately using the Gravitational Wave Interferometer Noise Calculator (GWINC). The red line, labeled "Mirror Coating Brownian" is one of the limiting noise sources at mid-frequencies, and is described by Equation 1.

Designs for future detectors take advantage of many of the parameters in Equation 1. The planned KAGRA, the Einstein Telescope¹⁷, and the proposed LIGO Voyager upgrade¹⁸, are all intended to be operated at cryogenic temperatures (reduced *T*), with larger beam spots (increased *w*), and with stiffer substrate materials (increased *E*). The ideal coating materials for these future detectors will have lower internal friction (reduced ϕ_{coat}), and thinner films (reduced *d*), and the methods for achieving such films has been an area of intense research for more than a decade. For the sake of this review, we will focus primarily on the reduction of ϕ_{coar} .

2. Mirror Film Structure and Optical Considerations

All but one of the currently-operating interferometric GW detectors use similar test mass mirror designs, which consist of a fused silica mass to act as both the test mass and the substrate for the mirror coatings. The one exception is that of the KAGRA detector, which uses sapphire substrates and cryogenic operation. The coatings themselves are multi-layer dielectric coatings composed of alternating high-refractive-index, n_H, and low-refractive-index, n_L, amorphous layers deposited using Ion Beam Sputtering (IBS). Dielectric mirror stacks of this type are used primarily to reach the interferometers' stringent optical requirements¹. For example, the Advanced LIGO detector requires mirror coatings with optical absorption of less than 0.5 ppm, optical scatter of less than 10 ppm, and a surface figure deviation of less than 0.7 nm RMS19. To date, no coating vendor has been able to match these requirements using polycrystalline films or other deposition methods.

The mirror coatings are composed of silica (SiO_2) as the n₁ material and titania-doped tantala $(Ti:Ta_2O_5)$ as the $n_{\rm H}$ material. IBS coatings of metal-oxides such as these are often found to be oxygen-poor, which contributes to increased optical absorption in as-deposited films²⁰. This is remedied through annealing the films in ambient air to temperatures as high as 600° C. For general-purpose high-reflectivity mirrors, the optical thickness, n^*l , where *n* is the refractive index and *l* is the physical thickness, of the individual layers is chosen to be 1/4 of the wavelength of the reflected light, and the number of layer pairs determines the total reflectivity of the coating. The test-mass mirrors used in Advanced LIGO have been slightly modified from this design, and have been optimized for reflecting light at two different wavelengths while minimizing material contributions to the value of ϕ_{coat}^{1} .

3. Loss Measurement Techniques

Within the GW community, the most common method for determining the internal friction of thin films is by applying those films to well-charactarized substrates with extremely low internal friction and then measuring the change in mechanical quality factor, Q, of the substrate's resonant modes. Absent any external losses, the internal friction of the film, ϕ_{film} , is related to Q of the coated substrate, Q_{coated} by:

$$Q_{coated}^{-1} \simeq Q_{substrate}^{-1} + \frac{U_{film}}{U_{total}} \phi_{film}, \qquad (2)$$

where $Q_{substrate}$ is the Q of the uncoated substrate at the same resonant mode, and U_{film}/U_{total} is the ratio of elastic energies stored in the film to the total energy in the combined film/substrate system at the resonant mode. Here, we use $\phi_{_{film}}$ instead of ϕ_{exect} in order to differentiate between the internal friction of an individual thin film under measurement and the internal friction of the complete multilayer coating used for making GW detector mirrors in Equation 2. In practice, the value of Q^{-1}_{coated} is often more than an order of magnitude greater than $Q^{-1}_{substrate}$, despite the fact that $U_{\rm film}/U_{\rm total}$ is generally much less than 10⁻³, due to the much higher value of $\phi_{\rm film}$. We also highlight the difference between Q, a measure of all of the mechanical losses at the substrate's resonant frequency, and $\phi_{\rm film}$, a derived value of the film's internal friction, which in this case is measured at the resonant frequency of the substrate.

For room temperature measurements, the substrate is generally made of fused silica, similar in quality to that of the test mass mirrors in GW detectors. The substrates are made in the shape of a 3-inch (~7.6 cm) diameter disc, with a thickness on the order of 1-2.5 mm. The discs can be suspended using a welded-silica thread to minimize energy loss from the vibrational mode of the disk into the support structure^{15,21-23} or balanced at a nodal point of the resonant modes²⁴⁻²⁶. The resonant modes of the disc are excited using an electrostatic excitation plate, and the oscillation of the mode is read using either a birefingence sensor or optical lever; both of these methods exert negligible back-action.

Once a mode is excited, the driving force is removed, and the Q can be determined by the ring-down timescale of the oscillation. The value for U_{film}/U_{total} is calculated using finite element modeling, and depends upon the elastic properties of the film under study, as well as its thickness relative to the substrate thickness²⁷. The internal friction of the coatings used in the Advanced LIGO GW detectors have been estimated from measurements made using these techniques, and the loss of the complete mirror stack has been calculated to be roughly 1×10^{-4} ¹⁵. This value has been roughly verified through direct measurements of S_{CBTN} in laboratory interferometers^{28,29}. It has been found that this mechanical loss is dominated by the tantala layers, and the interfaces do not contribute significantly to the loss²².

For measurements at cryogenic temperatures, the substrates of choice are silicon cantilevers, usually 0.5-1 cm in width, 4 cm long, and 50 µm thick, manufactured so that there is a thicker (~500 µm) clamping block on one end³⁰⁻³³. The clamping block is held between two stainless steel blocks mounted within a cryostat, where the temperatures are generally controlled between 10 and 300 K. The bending modes of the cantilever are excited with an electrostatic drive plate, and the oscillations observed with either an optical lever or a shadow sensor. The cryogenic Qs of these substrates can reach $\sim 10^7$, presumably limited by clamping losses, and decrease steadily above 100 K due to thermo-elastic loss, ultimately reaching values of ~104 at room temperatures³⁰. The value for U_{film}/U_{total} for the bending modes of a thin cantilever coated on one side can be calculated using the equation,

$$\frac{U_{film}}{U_{total}} = \frac{E_{substrate} t_{substrate}}{3E_{film} t_{film}},$$
(3)

where *E* is the Young's modulus, *t* is the thickness, and the subscripts _{substrate} and _{film} refer to the substrate and film, respectively³¹.

4. Methods for Reducing the Mechanical Loss of IBS Optical Films

At room temperatures, research within the GW community has discovered two methods for reducing the internal friction of IBS silica and tantala films. The first method is that of annealing. As deposited, silica films have an internal friction in the low-10⁻⁴ values. Annealing can reduce this value by almost an order of magnitude³⁴, with higher annealing temperatures leading to lower internal friction, ultimately limited to the loss value of the surface loss of bulk samples^{35,36}. A similar trend is seen in the tantala layers, where internal friction is reduced, although less dramatically, with increased annealing temperature until the material crystallizes above 600° C³³. Work is underway to increase this crystallization temperature in tantala and other materials through the use of doping and nano-layer deposition^{37,38}. Studies of structural changes in tantala show that this decrease in internal friction is correlated with increased medium-range order³⁹.

The second method for reducing room-temperature internal friction in tantala is through the addition of titania (TiO₂) doping. The room temperature internal friction of un-doped and un-annealed tantala films is in the high 10^{-4 22,21}. By doping the materials with >20% titania, this loss can be reduced by as much as 40%²³. This doping has the added advantage of increasing the value of n_{μ} , allowing for the reduced amount of material in the films, and further reducing the value of ϕ_{and} and d. Atomic structure measurements show that the reduction in internal friction is correlated with increased short-range order within the material⁴⁰. Atomic modeling and analysis of the cryogenic internal friction indicates that there is an increase in activation energy of the associated loss mechanisms^{41,32}. Other dopants have been explored, but to date, no combination has given a better film than titania-doped tantala42.

Cryogenic measurements of the internal friction of IBS silica and tantala films show a worrying trend for future cryogenic GW detectors, in that both materials exhibit a peak in mechanical loss around 20 K. In silica, this loss peak reaches values in the upper-10-4 36. While annealed tantala and titania-doped tantala films can reach as high as 10^{-3 31-33}. If these films were used in a GW detector operating at 20 K, the reduction in S_{CBTN} due to the lower value of T (see Equation 1) would be counter-acted by the increased value of ϕ_{con} , reducing the benefit by a factor of 2³⁸. Contrary to trends seen in room-temperature internal friction measurements, the loss peak in tantala appears to grow in response to higher annealing temperatures^{32,33}. In amorphous materials, these loss peaks are generally associated with thermally-excited Two-Level Systems (TLS). Two level systems are small configurations of atoms within the material where there exist two local configurational energy minima separated by a small activation energy⁴³. In silica, the activation energy for the most prominent loss peak is about 32 meV³⁶ while in pure tantala, it was measured to be 28.6 meV32 and in titania-doped tantala, this value is increased to 42 meV³³.

5. Current Research Directions

Atomic modeling efforts are underway to identify the physical nature of TLS in optical thin films^{41,44-46} with the goal of computationally predicting coating materials that can have a low internal friction, thereby reducing the number of physical films to be measured in the laboratory. In general, it has been discovered that TLS are a broad population of mechanisms involving bond rotations and reformations involving a few to tens of atoms throughout the material. Recent work has elucidated the effects of dopants on the population of TLS⁴¹ and shown how loss peaks at higher temperatures may be explained by separate populations of TLS within the material⁴⁶.

Another quickly-growing research direction within the GW community is the exploration of TLS-free materials, which would have drastically-reduced internal friction at cryogenic temperatures. Work done at the Naval Research Laboratory, in collaboration with Berkeley, has shown that amorphous silicon (a-Si) e-beam evaporated upon heated substrates exhibits no TLS47-49. This method of film deposition is similar to that of Ultra-Stable Glasses (USG) known within the organic glass community50. These a-Si films exhibit many similarities with USG, including increased density and reduced heat capacity. Another USG, indomethacin $(C_{10}H_{16}CINO_4)$, has also been shown to have no TLS⁵¹. This has led to the exploration of methods for making USG forms of common optical materials for use in GW detectors. Recent work within the Naval Research Laboratory has shown that it is possible to make low-TLS a-Si using magnetron sputtering, which produces films with higher-densities than e-beam evaporation, and may possibly reduce the need for high substrate temperatures. This can be seen in Figure 2. TLS-free a-Si exhibits optical absorption greater than those required by GW detectors; however, the material may still be useful as a buried layer in multi-material coating designs⁵².



Figure 2. Internal friction of magnetron sputtered a-Si films deposited on substrates at elevated temperatures. The faded background plot is from reference Liu et al.⁴⁷, and shows similar measurements for e-beam evaporated films. Sputtered films show reduced loss at the same substrate temperatures, indicating that higher-energy deposition processes may require lower substrate temperatures to produce TLS-free films.

Just as an amorphous material requires two elastic parameters to fully describe the elastic response of the material (e.g., Bulk Modulus and Shear Modulus, Young's Modulus and Poisson ratio, etc.), two associated anelastic parameters are required to fully account for the internal friction of material (e.g., ϕ_{Bulk} and ϕ_{Shear}). Another recent discovery within the GW community is that the internal friction associated with bulk deformations, ϕ_{Bulk} , is different from internal friction associated with shear deformations, ϕ_{Share} , and that this difference gently hints at a frequency dependence⁵³, as can be seen in Figure 3. The ratio of ϕ_{Bulk} ϕ_{Shear} has importance to GW detection, as this value can impact the calculation of S_{CBTN}^{16} . To the best of the author's knowledge, this is the first ever measurement of this ratio for an amorphous material, and the frequency dependence has no theoretical explanation.



Figure 3. Measurements of Shear and Bulk internal friction in titania-doped tantala films showing a possible frequency dependence. These figures are taken from Abernathy et al.⁵³.

Finally, future GW detectors may transition to epitaxially grown crystalline coatings^{54,55}. These coatings are known to have internal friction values a factor of ten lower than those of currently-used amorphous coatings⁵⁴. A great detail of research is needed, however, to scale these coatings to match the size and optical requirements of GW detectors⁵⁶. As these materials have multiple crystalline symmetries, even more elastic and anelastic parameters are required to fully describe their thermal noise contributions; however, the repercussions of having more than two internal friction parameters are still under exploration.

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