

Redefining precision interferometry and spectroscopy with high-performance optical interference coatings

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Abstract: High-performance optical interference coatings have transformed precision interferometry and spectroscopy by enabling unparalleled control over light-matter interactions. This review explores recent innovations in ion-beam sputtered amorphous dielectric, as well as substrate-transferred crystalline coatings, and their impact on systems at the forefront of precision metrology. These state-of-the-art coating techniques generate multilayers with ultralow optical losses, yielding mirrors with exceptional reflectivity. Refinements in their noise performance push the ultimate limits of sensitivity, resolution, and stability in demanding laser-based metrology applications. These technologies underpin the most advanced timekeeping and spatial measurement tools, enabling high-finesse reference cavities for the world's most precise optical atomic clocks and low-noise reflective test masses for km-baseline gravitational-wave detectors. Emerging hybrid designs combining these techniques expand access to the mid-infrared spectral region, enabling the first ultralow-optical-loss coatings in the 3000 nm to 5000 nm wavelength range for enhanced spectroscopy and trace-gas detection. We highlight how these technologies redefine coating performance metrics and set new benchmarks in quantum science, fundamental

49 physics, and precision optical sensing.

50 1. Introduction

51 Multilayer interference coatings are a key component of a wide variety of optical systems,
52 integrated in consumer products, advanced engineering tools, and fundamental scientific ex-
53 periments. In their most basic form, these structures consist of alternating thin films of high
54 and low refractive index materials that enable the control of incident light via wave interference
55 effects. Careful layer design and thin-film materials selection enable the realization of essentially
56 arbitrary optical functionality. Select examples include antireflection (AR) coatings, which are
57 engineered to destructively interfere the incident and reflected field; multi-band and partially
58 reflective structures employed for dichroic mirrors or beam splitting applications; low-pass,
59 high-pass, and notch-based narrow band-pass filtering; as well as high-reflectivity (HR) coatings,
60 which rely on constructive interference to maximize the reflection of an incident light field [1].
61 For resonant optical cavities, high-performance HR coatings are an essential component. One
62 of the simplest forms of an HR multilayer is a distributed Bragg reflector (DBR). In contrast
63 to broadband metallic mirrors, DBRs exhibit a narrow region of high reflectivity at a desired
64 center wavelength (CWL), λ . These structures consist of alternating layers of high and low
65 refractive index materials with individual layer thicknesses corresponding to one quarter of the
66 CWL when scaled by the material refractive index, commonly referred to as a $\lambda/4$ -stack. The
67 behavior of such interference coatings is fully governed by linear superposition of fields reflected
68 and transmitted according to Fresnel equations. Details are illustrated in Fig. 1.

69 Transfer-matrix methods are required for detailed analysis of an optical interference coating,
70 but analytical methods [2] can be used for example to determine the relative width of the stopband,
71 as set by the refractive-index contrast,

$$\frac{\Delta f}{f_0} = \frac{4}{\pi} \sin^{-1} \left(\frac{n_H - n_L}{n_H + n_L} \right) \quad (1)$$

72 with n_H and n_L being the high and low refractive index components of the DBR respectively.
73 The maximum reflectivity further depends on the number of mirror periods (alternating double
74 layers) employed, N , as well as the incident and substrate refractive indices, n_i and n_s , with the
75 expression below for an ideal quarter-wave multilayer at normal incidence [3]

$$R = \left[\frac{n_i n_H^{2N} - n_s n_L^{2N}}{n_i n_H^{2N} + n_s n_L^{2N}} \right]^2. \quad (2)$$

76 The performance of such a mirror is captured by three components, its transmission, T ,
77 absorption loss, A , and scatter loss, S , with the reflectivity, $R = 1 - (T + A + S)$. In this
78 case, the transmission is a design parameter, dependent on the refractive index contrast of the
79 component materials (difference between the material indices), as well as the total thickness
80 of the structure driven by the corresponding number of alternating layer pairs. Typical DBR
81 designs range from only a few layer pairs to several tens of periods, depending on the multilayer
82 refractive index contrast, enabling tunable transmission values from the percent regime down to
83 the parts-per-million (ppm, 10^{-6}) level at the CWL. With optimized materials and manufacturing
84 techniques, it is possible to reduce the excess optical losses, L , the combined optical power
85 absorption and scattering losses, $L = A + S$, to comparable ppm levels. Mirrors with such ultralow
86 excess losses ($L < 10$ ppm) are herein referred to as "supermirrors" regardless of the materials
87 or deposition technique used. Such devices are realized by minimizing intrinsic absorption
88 of the constituent materials, driven by background impurities or other optically-active defects,
89 non-linear absorption processes at high intensities, as well as by minimizing optical scatter, which

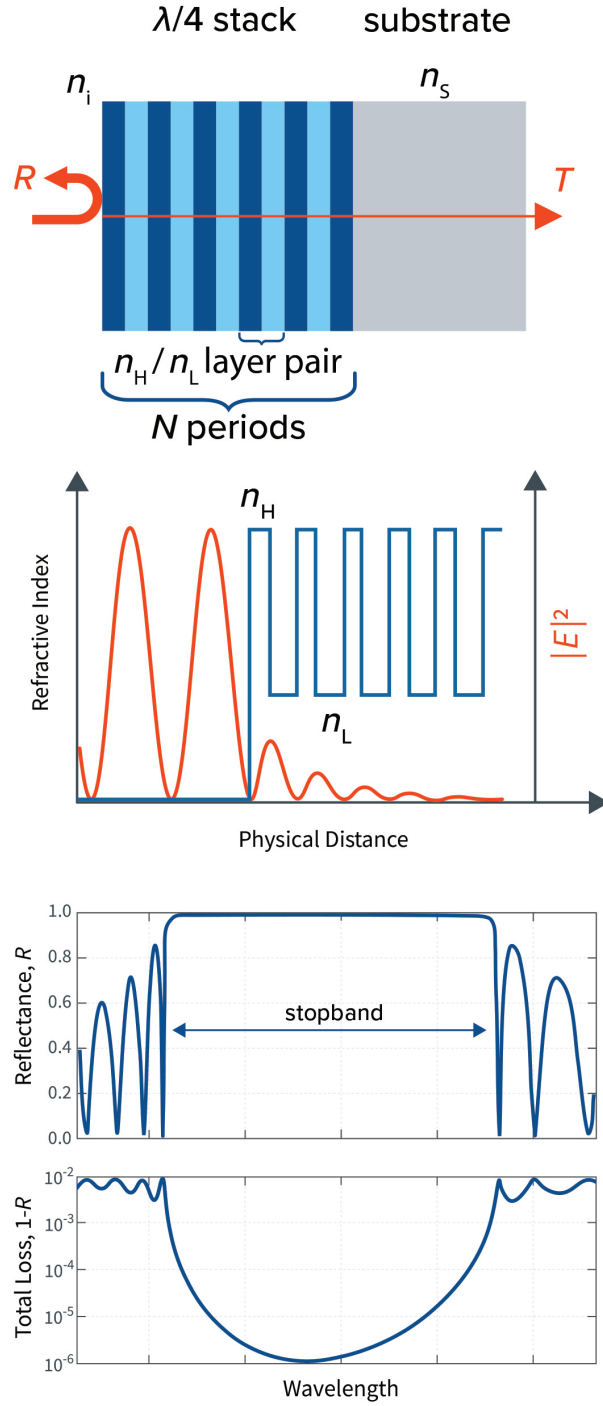


Fig. 1. *top panel:* Schematic of a quarter-wave ($\lambda/4$)-stack, a standard design for high-reflectivity mirrors, comprising N periods comprising alternating layers of high-index (n_H) and low-index (n_L) materials deposited on a substrate of refractive index n_s , the thick and thin arrows indicating reflected (R) and transmitted (T) light, respectively. *middle panel:* Plot of the refractive-index profile and the field strength of the standing wave formed by reflection from the interference coating for an incident wave in a medium with refractive index n_i . *bottom panel:* Calculated reflectance, R , of an ideal $\lambda/4$ -stack and the corresponding total loss ($T + L = 1 - R$), highlighting the characteristic high-reflectivity band centered around the design CWL.

necessitates excellent surface quality (i.e., sub-nanometer root-mean-square microroughness), a low structural defect density, and smooth texture-free interfaces.

In resonant cavity systems, the excess optical loss defines the on-resonance transmission T_{cav} [4], controlling the throughput or contrast, and also the signal-to-noise ratio (SNR) in cavity-enhanced spectroscopy efforts, according to $T_{\text{cav}} = T^2/(T + L)^2$, assuming equal transmission for both mirrors of a linear cavity and perfect mode matching. Here, one aims to maximize the cavity finesse, $\mathcal{F} = \pi/(T + L)$, which is inversely proportional to the cavity total loss, $T + L$. L is ultimately limited by the choice of materials and manufacturing technology. If L is non-negligible in comparison to T , it is necessary to strike a compromise in the above figures of merit [5].

Coating solutions capable of ultralow excess optical losses were originally developed for the construction of HeNe-based ring-laser gyroscopes in the late 1970s [6] and employed ion-beam sputtered (IBS) transition-metal-oxide multilayers. This technology, developed by Litton Guidance and Control Systems for military and aerospace applications, marked a turning point in the transition from mechanical to solid-state inertial navigation systems. Continuing to push the limits of optical performance, Rempe et al. [7] reported an IBS coating at 850 nm on conventionally-polished substrates with transmittance $T = 0.5$ ppm and excess loss $S + A = L = 1.1$ ppm. High-quality near-infrared (NIR) mirrors are now routinely capable of ppm-level excess optical losses [8, 9], and recently IBS coatings on micro-fabricated mirrors have realized record-low excess losses of $S + A = 0.74$ ppm (with $T = 1.9$ ppm) at 1550 nm [10, 11] enabling $\mathcal{F} > 1 \times 10^6$. These performance levels have been realized in the NIR for both IBS and crystalline coatings, and recently for all crystalline and hybrid mirrors (combining both epitaxial and IBS films) in the mid-IR (MIR), for coating CWLs near 4500 nm [12, 13].

Thus, in the NIR, from roughly 1000 nm to 2000 nm, optical performance has been pushed to its effective limits. However, for precision optical metrology applications, which represent the leading-edge of performance requirements for such coatings, a high-finesse optical cavity alone is not enough. Interferometric systems rely on extremely accurate sensing of changes in phase of a probe laser beam, which in turn requires high-stability cavities, entailing the simultaneous minimization of optical and *elastic* losses, particularly for applications in optical atomic clocks and gravitational-wave detectors (GWDs), as discussed in detail below. Another topic of great interest involves extending the wavelength range for supermirrors with ppm-level optical losses to the MIR and beyond for optical enhancement and ring-down cavities. These precision amplitude measurements have utility for advanced spectrometers probing various rotational-vibrational modes of molecules for trace gas detection. In this review we take a deep dive into these topics, exploring progress to date and the path forward in reducing the deleterious effects of thermal noise in optical interference coatings, as well as pushing the limits of optical loss in long-wavelength cavity end mirrors. We restrict our review to leading candidate technologies in these domains, including IBS metal oxide systems and substrate-transferred epitaxial coatings based on GaAs/AlGaAs multilayers. While other technologies are also promising, they are less mature. For example, amorphous semiconductors such as a-Si and semiconductor-derived dielectrics such as SiN_x are under active investigation, their inclusion here is necessarily limited, as they are not yet broadly implemented in the primary application domains emphasized in this review. Future work will center on advanced materials synthesis and optimization of production methods, exploring alternative coating materials and combinations thereof, from amorphous oxide mixtures to novel single-crystal systems including III-phosphides, III-nitrides, as well as epitaxial oxides.

2. Low-Loss Optical Interference Coatings

2.1. Synthesis of high-performance amorphous coatings

Amorphous, commonly referred to as "dielectric", interference coatings are synthesized by physical vapor deposition (PVD) processes, i.e., ion beam and magnetron sputtering, as well as

ion-beam-assisted evaporation [14]. For demanding precision optical metrology applications in the visible and NIR spectral regions, dielectric coatings based on amorphous metal oxide multilayers are fabricated primarily by IBS. This technique has produced mirror coatings for GWDs with the lowest excess optical losses reported to date and thus remains the preferred process solution [9].

A schematic of the IBS process is shown in Fig. 2. In a gridded source system, an ion source produces a beam of energetic noble gas ions that sputters a target, either metal or oxide in an oxygen-rich atmosphere. This creates a plume of atoms and oxide molecules which impinge on the surface of a substrate and stick at this location due to limited mobility. IBS thin films are amorphous, stoichiometric, and uniform, being highly densified and exhibiting compressive stress with extremely low roughness (and thus minimal optical scatter), as well as high purity for minimal absorption. In a typical HR design, SiO_2 is used for the low refractive index component, in combination with a high-index transition metal oxide such as Ta_2O_5 , TiO_2 , and HfO_2 .

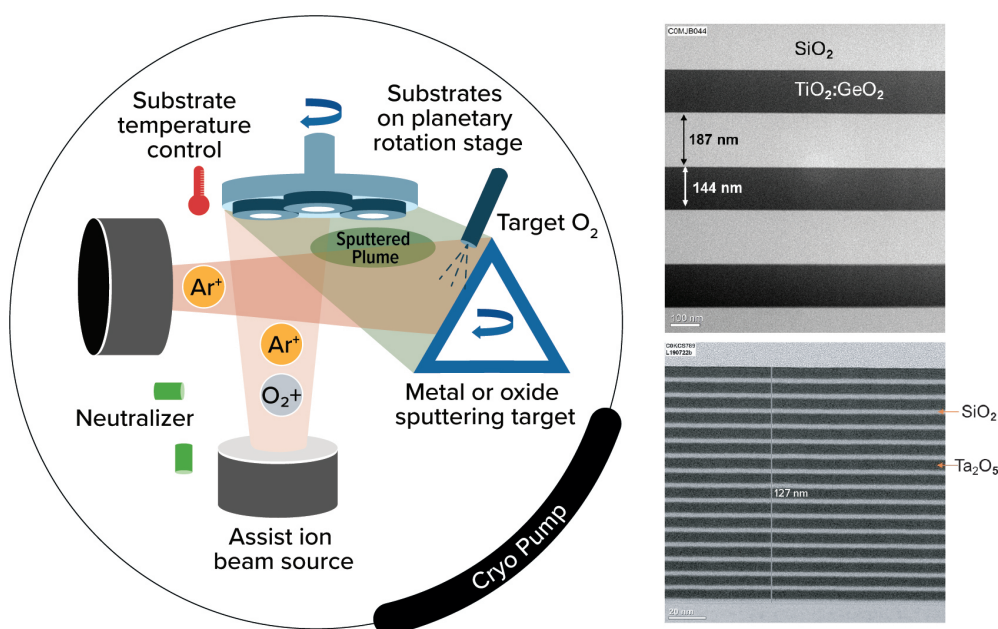


Fig. 2. *Left*: Schematic of a dual ion beam sputtering system. The main ion source creates a beam of noble gas ions that sputters a target. The plume of atoms and molecules that is created deposits onto the substrates. The process takes place in an oxygen rich atmosphere. A secondary ion beam source operated with Ar^+ or O_2^+ can be used to modify the growing film. The system allows for substrate temperature control. Deposition rates are a few $\text{\AA}/\text{sec}$, therefore there is high control of the layer thickness. *Right, top*: Bright field transmission electron microscope (TEM) image of an amorphous $\text{TiO}_2:\text{GeO}_2/\text{SiO}_2$ multilayer dielectric coating showing extremely sharp interfaces. *Right, bottom*: TEM image of a 127 nm thick $\text{Ta}_2\text{O}_5/\text{SiO}_2$ nanolaminate.

Beyond binary compounds, amorphous oxide mixtures can be obtained by co-sputtering two different targets. There are many process parameters, from the ion source current and voltage, oxygen and argon (or other noble gas) flow, and chamber base pressure that are controlled to realize stoichiometric thin films with strong adhesion to the base optical substrate. The use of mixtures offers multiple benefits: high crystallization temperature [15], low optical losses, and stress control [16]. Amorphous oxide nanolaminates, consisting of alternating nanometer-thick layers of a high-index amorphous oxide and SiO_2 with an effective medium refractive index

159 similar to mixtures have also been proposed to improve optical and elastic losses [17]. In the case
160 of dielectric coatings for GWDs, annealing to temperatures near the crystallization point of the
161 high-index material, $\sim 600^\circ\text{C}$ is necessary to realize the lowest absorption and elastic losses [18].

162 2.2. Leveraging semiconductor infrastructure for crystalline coatings

163 The first demonstration of an epitaxial optical interference coating was carried out by J. P. van
164 der Ziel and M. Ilegems at Bell Labs in 1975 [19], coinciding with the initial development
165 of low-loss IBS coatings by Litton Industries as introduced above. In contrast to the utility
166 of PVD for the production of amorphous coatings, the growth of high-quality epitaxial films
167 has the distinct *disadvantage* of being constrained by lattice matching requirements. Here,
168 the growth of a low-defect-density single-crystal structure is limited to the deposition of thin
169 films on a planar surface with a nearly identical inter-atomic spacing, as well as a matched
170 crystalline symmetry. Thus, compound semiconductor interference coatings were confined to
171 the domain of surface-normal optoelectronics, notably as a key component in vertical-cavity
172 surface-emitting lasers (VCSELs). VCSELs are a unique class of microcavity lasers, comprising
173 a multi-quantum-well active region sandwiched between two epitaxial DBRs and are now the
174 largest volume laser produced. Commercial applications span facial recognition systems in smart
175 phones to optical mice, disk read systems, and high-speed datacom applications [20].

176 Roughly two decades ago, the simultaneously low optical and elastic losses observed in
177 suspended optomechanical resonators sculpted from GaAs/AlGaAs drove the use of epitaxial
178 multilayers as a low-noise coating solution [21–25]. The high purity and near structural perfection
179 of these monocrystalline DBRs yields ppm-level excess losses, and more importantly a 10- to
180 100-fold reduction in elastic losses. For crystalline microresonators, this enabled landmark
181 demonstrations including broadband quantum radiation pressure noise at room temperature
182 [26], ponderomotive squeezing [27], tailoring quantum noise via squeezed light injection [28],
183 back-action-evading measurements [29], and the realization of displacement sensitivity below
184 the standard-quantum-limit [30]. More practical applications followed, including laser power
185 stabilization [31, 32] and enhanced quantum-limited interferometry via optical spring tracking
186 [33]. Leveraging these unique properties, substrate-transferred GaAs/AlGaAs coatings were
187 first demonstrated in 2013. A substrate-transfer coating process was developed to integrate
188 epitaxial DBRs with arbitrary optical substrates as a means to reduce Brownian coating thermal
189 noise (CTN) in precision interferometry. The mirror performance was vetted by constructing a
190 cavity-stabilized laser system, with $\mathcal{F} \approx 150\,000$ at 1064 nm, and, most importantly, a reduction
191 in the coating elastic loss by a factor of 10 compared to amorphous oxide coatings [34].

192 In the crystalline coating process, lattice matching constraints between the epitaxial films (i.e.,
193 the crystalline coating layers) and the substrate are overcome by using a multi-step manufacturing
194 process. This combines established techniques relevant to semiconductor microfabrication
195 spanning epitaxial growth, lithography, selective etching, and plasma-activated direct bonding
196 [35], as detailed in Fig. 3 below (additional details included in a Supplementary Document).
197 This process allows for the flexible and *facile* integration of epitaxial coatings onto essentially
198 arbitrary optical substrates, currently up to a maximum diameter of 200 mm, limited by the largest
199 currently available GaAs based wafers [36]. This can be increased to 300 mm by exploiting
200 heteroepitaxial growth of GaAs/AlGaAs multilayers on Ge substrates [37, 38]). One key aspect
201 of the process is the ability to realize single-crystal coatings on curved surfaces, with a current
202 minimum radius of curvature of 5 cm [39]. While fused silica is the most common substrate
203 material for room temperature and NIR optics, high quality crystalline GaAs/AlGaAs coatings
204 have been realized on a wide variety of substrates including single-crystal Si (c-Si), sapphire,
205 SiC, diamond, YAG, Ge, and YVO₄ among others.

206 Further advancements in substrate-transferred crystalline coatings focus on pushing the limits
207 of purity, surface quality, and maximum area, thereby bringing the unique properties of these

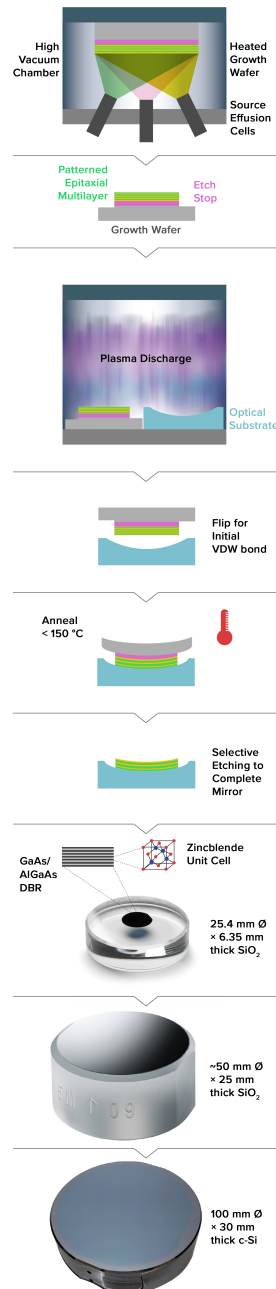


Fig. 3. Overview of the substrate-transfer crystalline coating process and example crystalline mirrors. This multi-step procedure involves, from top to bottom: MBE growth of the epitaxial optical interference coating on a lattice-matched base wafer. Lithographic patterning and either wet or dry (plasma-based) etching to define the coating geometry. Cleaning, plasma activation, and inspection of both the epiwafer and optical substrate immediately ahead of the bonding process. Flipping the epiwafer and pressing into contact with the optical substrate, applying controlled pressure, initiating a van der Waals (VDW) bond. Annealing the stack to strengthen the bond and enhance the interfacial energy. Wet chemical selective substrate and etch stop removal, followed by final cleaning, and inspection of the completed crystalline mirror. The final three panels include photographs of: i) a typical 1" diameter reference cavity mirror with an 8-mm diameter epi coating disc on a fused silica substrate. ii) Photo of a custom optic consisting of a 48.8 mm diameter crystalline coating on a 5-m concave radius of curvature silica substrate. iii) Photo of a 100 mm diameter custom crystalline mirror on a c-Si substrate.

materials to bear on demanding precision laser optics applications. With continuous refinement over the last decade, excess optical losses have been demonstrated to be < 5 ppm (with absorption < 1 ppm confirmed in the NIR [40]), enabling $\mathcal{F} > 400\,000$ for center wavelengths spanning 1064 nm to 4500 nm [12]. In the NIR, the performance of crystalline coatings is on par with high-quality IBS films, while in the MIR, they are superior. These results confirm that an optimized MBE process is capable of producing extremely low excess optical losses. However, the limited electronic bandgap (1.4 eV or roughly 870 nm at 300 K) of the high-index GaAs layers precludes transparent operation in the visible spectral range, where IBS coatings currently excel. The exceptional IR optical performance of crystalline mirrors is complemented by additional advantageous physical properties including a low elastic loss, to be discussed in the following sections, as well as a reasonably high thermal conductivity ($> 20 \text{ W m}^{-1} \text{ K}^{-1}$) when compared with traditional sputtered metal oxide films ($\sim 1 \text{ W m}^{-1} \text{ K}^{-1}$).

3. Thermal Noise in Optical Resonators

After five decades of development, high-performance interference coatings are now indispensable for use in long-coherence-length lasers for optical atomic clocks, GWDs, and tests of fundamental physics [41]. In these applications, the mirrors are now running up against fundamental limits in terms of their opto-*mechanical* performance. Here, thermal equilibrium fluctuations result in modifications of the optical path length and hence unavoidable displacement noise [42–47].

While there are several thermally-driven noise processes, the most direct is Brownian CTN. Here, non-resonant, mechanical fluctuations of the mirror surface are driven via *elastic loss*. Another important thermally-driven noise process is thermo-optic (TO) noise, which encompasses both thermoelastic and thermorefractive effects. With TO-noise, equilibrium fluctuations of the local coating temperature induce optical path length changes via the thermal expansion coefficient, α and thermo-refractive coefficient, β [48, 49]. These thermally-induced changes to the coating thickness and to the refractive index alter the optical path length, but with opposite sign. Thus, by careful design of the coating, TO-noise can be eliminated to first order. In AlGaAs/GaAs coatings, where TO-noise is large compared to amorphous oxides, the requirement for cancellation is stricter and has been previously demonstrated [50]. Similarly, equilibrium fluctuations of the local temperature of the mirror substrate in the area of the cavity mode changes the path length via the substrate’s thermal expansion coefficient α_s , leading to substrate thermo-elastic noise. Other thermally driven noises, including substrate or spacer Brownian noise and noise due to rubbing at interfaces may also be important in certain situations. For calculation of reference cavity noise, an online calculator is available [51]. Together, these thermally driven noise sources set the ultimate stability limit in precision metrological systems [41].

Expanding on the Brownian CTN topic, the magnitude of Brownian noise is quantified via the frequency-dependent noise power spectral density, $S_x(f)$, and depends directly on the elastic loss of the component materials. Specifically,

$$S_x(f) \propto \frac{k_B T}{f} \frac{\phi}{w^2} \quad (3)$$

where k_B is the Boltzmann constant, T is temperature, f is Fourier frequency in the low-frequency limit far from any mechanical resonances of the system, w is the beam radius ($1/e^2$ intensity), and ϕ is the elastic loss angle of the optical coating. The elastic loss angle is the imaginary component of a complex Young’s modulus corresponding to the deformation consideration, $E(f) = E_0[1 + i\phi(f)]$, $E_0 \in \mathbb{R}$, and it is the fraction of elastic energy dissipated per cycle compared to the cycle-maximum elastic energy. The $1/f$ slope of the Brownian noise $S_x(f)$ leads to a laser frequency stability, expressed as Allan deviation, that is independent of averaging time, whereas other noise sources such as coating TO noise show different frequency dependencies and contributions to the stability that depend on averaging time.

In a resonant cavity, the total Brownian noise is determined by the elastic loss angle contributions of all materials making up the reflective elements, as well as the supporting structures for the system. In a reference cavity this would be the optical coating, the mirror substrate, and the rigid spacer that the mirror substrates are contacted to; in a GWD this would encompass the coatings and substrates of the reflective test masses, as well as the suspension system. From a conceptual viewpoint, for all of these components, at a finite temperature, the constituent atoms fluctuate around their fixed mean position leading to random Brownian noise [52]. While the equipartition theorem specifies that each degree of freedom (however constructed, e.g., of the normal modes) should carry an average $\frac{1}{2}k_B T$ of energy, their spectral distribution is described by the elastic loss angle $\phi = \Delta f / f_0$, where f_0 is the normal mode frequency and Δf is the FWHM of the mode. Thus the elastic loss, or material dissipation, ϕ , generates thermal noise at frequencies outside the resonance. This relation is the essence of the fluctuation–dissipation theorem [42–44] and its application to optical interferometry [45–47, 53, 54].

While we use Eq. 3 for this review, we note that this expression is only approximate and assumes a frequency-independent, homogeneous, and isotropic loss angle. In reality, the loss angles corresponding to bulk and shear moduli may be different, even in amorphous media. In anisotropic crystalline solids, there will be independent loss angles for each component of the elasticity or stiffness tensor. For a material with cubic symmetry such as GaAs, there are three loss angles ϕ_{11} , ϕ_{12} , and ϕ_{14} corresponding with the three elastic constants C_{11} , C_{12} , and C_{14} [55]. Combining both the high and low refractive index components of a GaAs/AlGaAs DBR, this yields six distinct loss angles considering the averaged properties of the multilayer as those of a single effective medium.

Thermal noise in optics, especially Brownian CTN, has now become a significant impediment to improved performance in ultrastable resonators and GWDs. As modal decomposition is only efficient for simple geometries [56], the so-called direct approach pioneered by Levin [45] is employed. This technique applies the fluctuation-dissipation theorem to relate the thermal fluctuation of a mechanical quantity x to the mechanical dissipation, that occurs when the conjugate force is applied to the system. In an optical resonator, the length fluctuations, averaged over the laser mode are related to the loss, when an oscillating pressure distribution due to the laser intensity distribution is applied to the cavity. Because of their direct and immediate interactions with the optical field, the coatings contribute most to the Brownian noise followed by mirror substrate and spacer, provided sufficiently low-loss materials are chosen. At room temperature fused silica mirror substrates (loss $\phi \approx 10^{-8}$ [57, 58]) and ultra-low expansion (ULE) glass or ceramic spacers are usually chosen for their low Brownian noise, however TO noise from the mirror coating and substrate can also become significant. For cryogenic systems, crystalline substrates and spacer materials including c-Si and sapphire are preferred, owing to excess elastic loss in amorphous systems.

Reducing CTN is now a crucial requirement for precision optical systems and requires simultaneous minimization of the elastic loss and optimization of the coating optical performance. Moreover, developing appropriate coating designs to minimize TO noise processes is key, particularly with crystalline coatings [50]. Further coating synthesis, materials, and design optimization will lead to resonant cavities with minimal thermo-mechanical fluctuations, reducing the noise floor and maximizing the system stability. To date, the implementation of ultralow-noise coatings has led to enormous progress (Fig. 4), yielding fractional frequency stabilities at the 10^{-17} level for state-of-the-art cryogenic reference cavities for stabilized laser systems employing crystalline coatings [59] and sub-attometer displacement sensitivities for km-baseline interferometric GWDs with large-area IBS coatings [60].

3.1. Impact on ultrastable lasers for optical atomic clocks

Optical Fabry-Perot resonators with rigid spacers act as frequency references for ultrastable lasers. These systems serve as interrogation oscillators in optical atomic clocks [61, 62], and are integral to high-precision tests of the equivalence principle [63, 64], and searches for dark matter [65, 66]. For these applications, the laser frequency stability is paramount to realizing improved system performance. Select examples include the realization of clocks with Dick-noise-limited frequency stability [62] and improved fidelity of qubit manipulations in quantum information science [67–69]. Understanding and mitigating fundamental noise sources that limit the frequency stability of cavity-stabilized lasers is critical for advancing these fields.

The fractional frequency stability (or equivalently, fractional length stability) of typical cavity-stabilized lasers is limited by CTN to approximately 10^{-16} , for averaging times between a fraction of a second to up to several thousand seconds. Fig. 4 provides a historical overview of progress in improving reference cavity stability over the last ~35 years. Mitigation strategies for Brownian thermal noise reduction in optical cavities is similar to that in GWDs, namely, enlarging the mode diameter on the mirrors and increasing the cavity length. Typical reference cavity mirror spacings range from a few millimeters to several tens of centimeters and beam diameters are commonly around 1 mm. Lowering the temperature of the cavity of course also helps to reduce the Brownian thermal noise, with mindfulness to material selection being key. Glassy systems exhibit loss peaks and degraded noise performance at low temperatures (for 100 K and below) [70, 71], while single-crystalline materials such as c-Si and sapphire show extremely low elastic losses in a cryogenic environment [72, 73]. However, impurities can also lead to loss peaks at low temperature [74, 75].

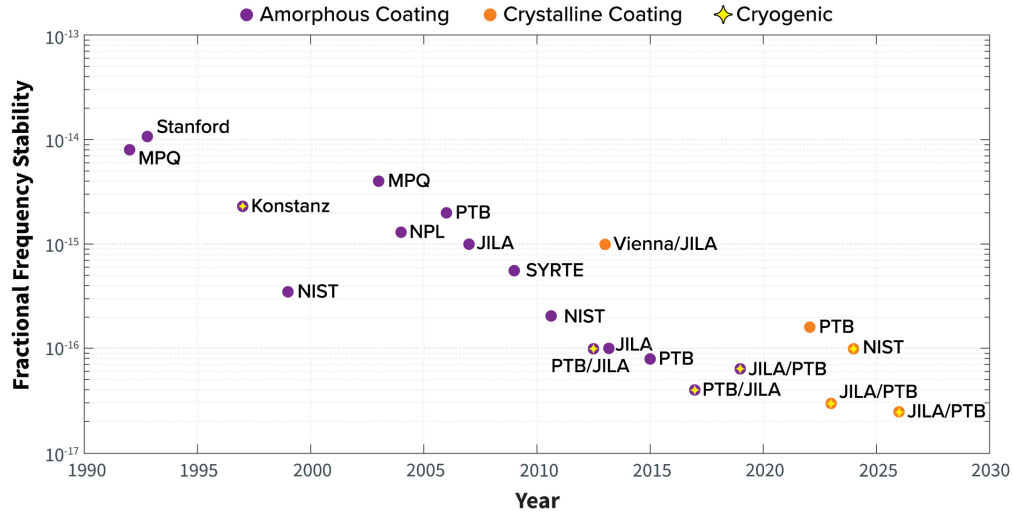


Fig. 4. Frequency stability achieved with cavity-stabilized lasers over 35 years. The labels refer to: MPQ 1992 [76], Stanford 1993 [77], Konstanz 1997 [78], NIST 1999 [79], MPQ 2003 [80], NPL 2004 [81], PTB 2006 [82], JILA 2007 [83], SYRTE 2009 [84], NIST 2011 [85], PTB/JILA 2012 [86], JILA 2012 [87], Vienna / JILA [34], PTB 2015 [88], PTB/JILA 2017 [89], JILA/PTB 2019 [90], PTB 2022 [91], JILA/PTB 2023 [92], NIST 2024 [93], JILA/PTB 2026 [59].

Beyond optimization of the spacer and mirror substrate materials, a key noise mitigation strategy is to search for coating materials exhibiting low elastic losses. Here, an important consideration for ultrastable cavities is that the mirrors must simultaneously exhibit exceptionally low optical losses to support a high cavity finesse (typically $\mathcal{F} > 100\,000$) and thus narrow

327 resonator linewidth. The latter is required to minimize the effects of photon shot noise and
328 electronic noise in the interrogation of the cavity. A sharper cavity resonance also mitigates
329 other technical noise contributions such as residual amplitude modulation associated with a
330 popular cavity locking technique, Pound-Drever-Hall [94]. Cavity linewidths of a few kilohertz,
331 achievable with a reasonable spacer length and ppm-level per-mirror optical loss, pushes the
332 shot noise contribution on the final laser stability to a negligible level for laser powers of a few
333 microwatts, and even down to ~ 100 nW coupled into the cavity [95].

334 The necessary few-ppm optical losses entails stringent requirements on the choice of mirror
335 coating materials, and is currently achieved in optimized IBS-deposited amorphous metal oxides
336 ($\text{Ta}_2\text{O}_5/\text{SiO}_2$ and related alloys), as well as MBE-grown substrate-transferred GaAs/AlGaAs
337 crystalline mirrors. For ultrastable reference cavities, these high-performance interference
338 coatings, both amorphous and crystalline, now enable the most advanced timekeeping instruments,
339 with recent examples of such room temperature coatings on fused silica for transportable clocks
340 [91, 96, 97] and future space platforms [98].

341 Cryogenic cavities, currently supporting the world's lowest-noise lasers, are well suited for
342 studies of coating noise. Due to the low elastic loss of c-Si, particularly at cryogenic temperatures,
343 the spacer and mirror substrates contribute negligibly, leaving the mirror coatings as the dominant
344 noise source. As one example, exploiting the vanishing thermal expansion of c-Si at 124 K, a
345 21 cm long resonator with IBS-deposited $\text{Ta}_2\text{O}_5/\text{SiO}_2$ coatings achieved thermal noise limited
346 instabilities of 4×10^{-17} for averaging times between 0.1 s and thousands of seconds [62, 89]
347 (referred to as "Si2", Fig. 5). A smaller, 6 cm long c-Si cavity at 4 K reached 6.5×10^{-17} [90].

348 Replacing the initially employed IBS coatings with substrate-transferred AlGaAs-based
349 crystalline mirrors (referred to as "Si5"), the lower elastic loss promises a reduction of the thermal
350 noise floor to about 1×10^{-17} . As the AlGaAs coatings exhibit static birefringence, the mirrors
351 were oriented with their fast optical axes in parallel to maximize the frequency splitting between
352 the orthogonal linearly polarized cavity eigenmodes. The differential phase shift of ≈ 1.1 mrad
353 on a single reflection between the two axis [36] lead to a splitting of 3.5×10^{-4} the free spectral
354 range of the resonator, $c/2L$, e.g., around 100 kHz for a 40 cm long cavity.

355 In experiments with reference cavities incorporating crystalline coatings, it was found that the
356 polarization eigenmode splitting can be further modified by the intracavity optical power, as well
357 as by above-bandgap illumination [100, 101]. Shifts on the order of kHz were observed with
358 an in-coupled laser power of 100 μW in a 48-cm room temperature cavity at PTB. Moreover,
359 illumination of the coatings with visible light led to a similar shift at intensities of 0.1 W/m^2 . The
360 observed slow temporal response rules out a thermal origin for this effect. To avoid frequency
361 noise arising from fluctuations of the intracavity power, the source must be sufficiently stable.
362 However, even with a well stabilized laser, spontaneous fluctuations of the splitting were observed
363 at 124 K, 17 K, and 4 K [92, 102, 103], leading to anti-correlated frequency fluctuations of
364 the orthogonally polarized cavity modes. As can be seen in Fig. 5, when a laser is frequency
365 stabilized to one mode of the Si5 cavity with crystalline coatings (Si5, Single Pol. dataset),
366 instabilities of 10^{-16} were observed, about a factor of ten above the expected Brownian noise
367 floor, the values of which can be seen in Fig. 6. As the fluctuations of orthogonally polarized
368 eigenmodes are anticorrelated, locking a laser to the average frequency of the two modes (Si5,
369 Pol. Ave. dataset), these birefringent fluctuations can be canceled.

370 Even with polarization averaging, first investigations of all-crystalline cryogenic reference
371 cavities at JILA and PTB revealed excess frequency fluctuations about a factor of four above
372 the expected Brownian noise floor, limiting the overall cavity performance [92, 103]. Noise
373 correlation between TEM_{00} and TEM_{01} modes indicated a spatial correlation length of the excess
374 fluctuations on the mirror surface larger than the mode size, very different from CTN, which
375 has a correlation length on the order of the coating thickness. Thus the processes were deemed
376 "global" excess noise. From correlation measurements, a coating loss angle, ϕ of $\approx 2.5 \times 10^{-5}$

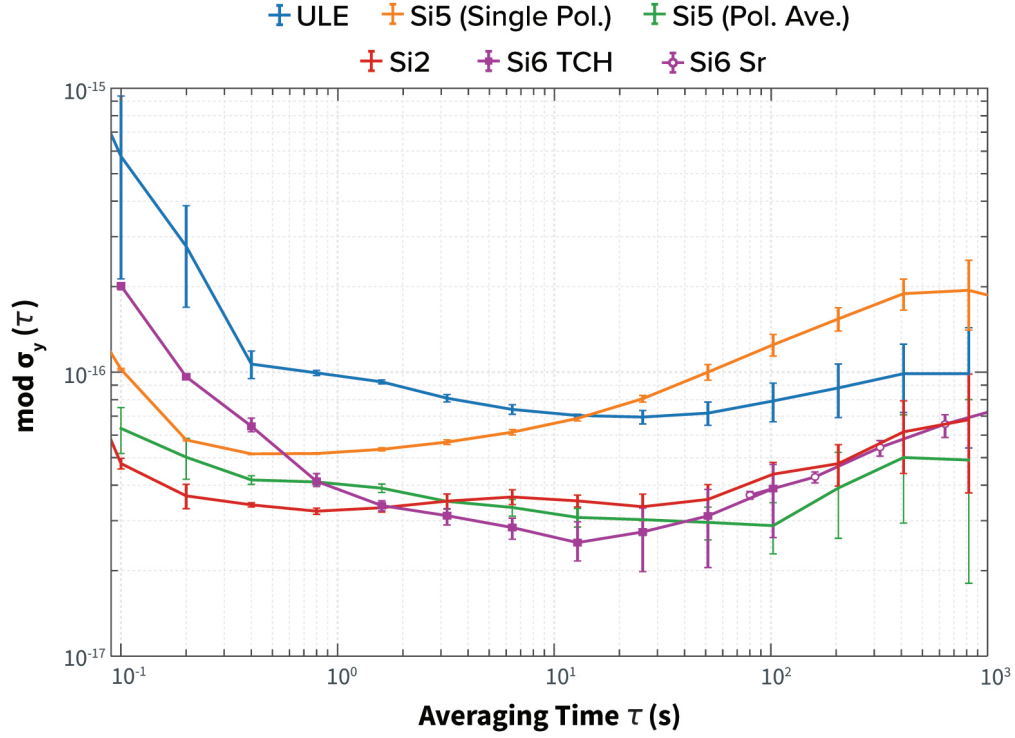


Fig. 5. Fractional frequency instability, expressed as modified Allan deviation, of a 48 cm long room temperature cavity (ULE) with dielectric coatings at 698 nm [88], a 21 cm long c-Si cavity at 124 K with amorphous mirror coatings (Si2), of an otherwise identical cavity with AlGaAs coatings (Si5) with single polarization lock and locking to the average frequency of the two polarizations, determined from a three-cornered hat analysis [99]. The uncertainty bars indicate the standard deviation of ten separate evaluations. In addition, the stability of a 6 cm long c-Si cavity with AlGaAs coatings at 17 K (Si6), determined by three cornered hat method at short averaging times and in comparison to a strontium lattice clock at longer times is shown [59].

could be determined in agreement with the values measured in GaAs/AlGaAs microresonators [21–25].

Following up, a second generation of AlGaAs-on-Si mirrors were supplied and recent measurements on the same 4 K / 17 K setup did not show this excess noise (Si6 cavity dataset in Fig. 5). Once again a dual locking scheme was employed to eliminate fluctuating birefringence, and, with a lack of global noise, the cavity instability was improved to 2.5×10^{-17} at 10 s of averaging [59]. The extracted value sets an upper bound to the 17 K loss angle of 2.3×10^{-5} , ten times lower than that measured for amorphous Ta₂O₅/SiO₂ coatings. This second generation of cryogenic crystalline mirrors were nominally identical to previous versions in terms of the epitaxial growth and substrate-transfer process. The only change was an increase in the number of layer pairs in the Bragg stack to reduce the optical transmission and enhance the mirror reflectivity. Thus, the excess or global noise observed previously does not appear to be inherent to crystalline coatings and remains under investigation.

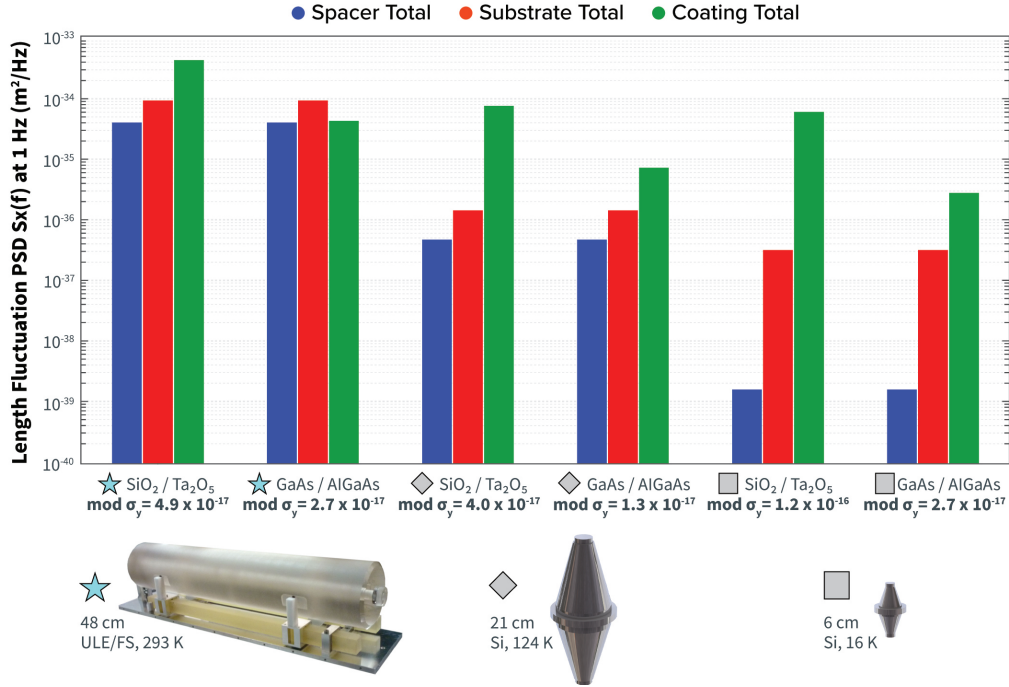


Fig. 6. Comparison of the power spectral density of length fluctuations S_x of spacer, substrate, and coating for glass- and c-Si-based cavities. The bottom panel includes photos or renderings of the cavities and indicates the cavity length, the spacer/mirror materials, and the operating temperature. The histograms at top show the noise contribution of the cavity components for each indicated coating type and are expressed as a corresponding modified Allan deviation $\text{mod } \sigma_y$ of the frequency. Note that this also includes the cavity length L ($\text{mod } \sigma_y^2 = 0.935 S_x(1 \text{ Hz})/L^2$, assuming $S_x \propto 1/f$ [104]). The length fluctuations of the room temperature ULE/FS cavities are calculated for the 48 cm long cavity design described in [88] at a wavelength of 1550 nm. The values for the c-Si cavities apply to realized systems at 124 K [89, 99] and 17 K [92, 105]. For the Ta₂O₅/SiO₂ coatings, a loss angle of 4×10^{-4} at 293 K [106], 2.4×10^{-4} at 124 K, and 3.2×10^{-4} at 17 K [105] are assumed. The loss angle of the GaAs/AlGaAs coatings is 2.5×10^{-5} .

3.2. Impact on GW astronomy

GWDs such as LIGO [107] are limited in their most sensitive frequency band by CTN of the from the amorphous-oxide coatings required to make the arm-cavity mirrors, known as the “test masses”, highly reflective. Fig. 7 shows an example of detector noise curves from the 4th observing run of LIGO, in this case for the Livingston Observatory, including an estimate of the thermal noise contributions. The most sensitive frequency band is the mid-band between about 40 Hz and 400 Hz. Also shown is an estimate of how the noise would be reduced if the current amorphous-oxide coatings were replaced with AlGaAs coatings. Even in current detectors, the sensitivity improvement from AlGaAs coatings would be significant. In future iterations of LIGO where the quantum noise is expected to be lower, the reduced CTN of AlGaAs complements the improved quantum noise, yielding significant mid-band sensitivity enhancements. It is noteworthy that the actual noise measured in the 4th observing run of LIGO was higher than the sum of all known noise sources. The reasons for this are not fully understood. However, the slope of the mid-band excess noise is consistent with the slope expected from CTN. Therefore, it is possible that CTN from the current amorphous oxide coatings is up to 30% higher than previously expected.

Currently, the coatings in LIGO are multilayer DBRs comprising IBS-deposited TiO₂ alloyed with Ta₂O₅ (titania-doped tantala), as the high-index layers, and SiO₂ as the low index films (TiO₂:Ta₂O₅/SiO₂). This coating composition was chosen for its outstanding optical properties, as well as reduced elastic loss compared with traditional Ta₂O₅/SiO₂ stacks. The alloyed high-index layers were developed after extending Levin’s groundbreaking paper on CTN [45], and measuring the elastic loss in these coatings. In these efforts, it was realized that existing coatings would not meet the Advanced LIGO requirements [106]. Further study determined that the excess elastic loss was due to the high-index Ta₂O₅ layers [109]. An exploration of amorphous coating materials determined that titania-doped tantala had a sufficiently low elastic loss and was a promising alternative. Its increased index reduced the total thickness of the high-index material, and by increasing the beam spot size, these coatings were able to meet the Advanced LIGO requirements [110]. To meet the stated scientific goals for the next upgrade for LIGO, it will be necessary to further reduce the Brownian CTN limit by at least 2× from the level shown by the “Thermal (AmOx)” trace in Fig. 7. If the proposed A[#] upgrade goes forward, then a reduction in thermal noise of at least 4× will be required. Currently, amorphous oxide coatings based on 44% Ti-doped GeO₂ (TiO₂ doped GeO₂) can achieve a 2× reduction in CTN [111], while AlGaAs coatings will enable a CTN that is at least 5× below the current level [112].

Analogous to efforts in high-performance reference cavities, a complementary approach to reducing thermal noise in GWDs is operation at cryogenic temperature. This strategy is currently implemented in the KAGRA detector [Akutsu], which employs sapphire test masses operated at ~20 K together with IBS amorphous oxide coatings [Hirose]. The use of sapphire is motivated by its high thermal conductivity and low mechanical dissipation at cryogenic temperatures, in contrast to fused silica. However, while cryogenic cooling reduces substrate thermal noise, amorphous coating materials exhibit increased mechanical loss at low temperature, typically with loss peaks in the 10-100 K range [113]. As a result, coating thermal noise remains a key limitation under these conditions, motivating the exploration of alternative coating platforms, such as crystalline multilayers, optimized for low-loss cryogenic operation.

3.2.1. Next generation amorphous oxides for future GWDs

Intensive empirical and theoretical studies by the GWD community have been devoted to addressing the requirements for next-generation HR amorphous oxide DBRs [9, 48, 52, 109, 110, 114–120]. One important observation is that the elastic losses of IBS silica (the current low refractive index material) are low enough that the high-index layer dominates the thermal noise, and the losses scale with the volume of that material rather than the number of interfaces [109].

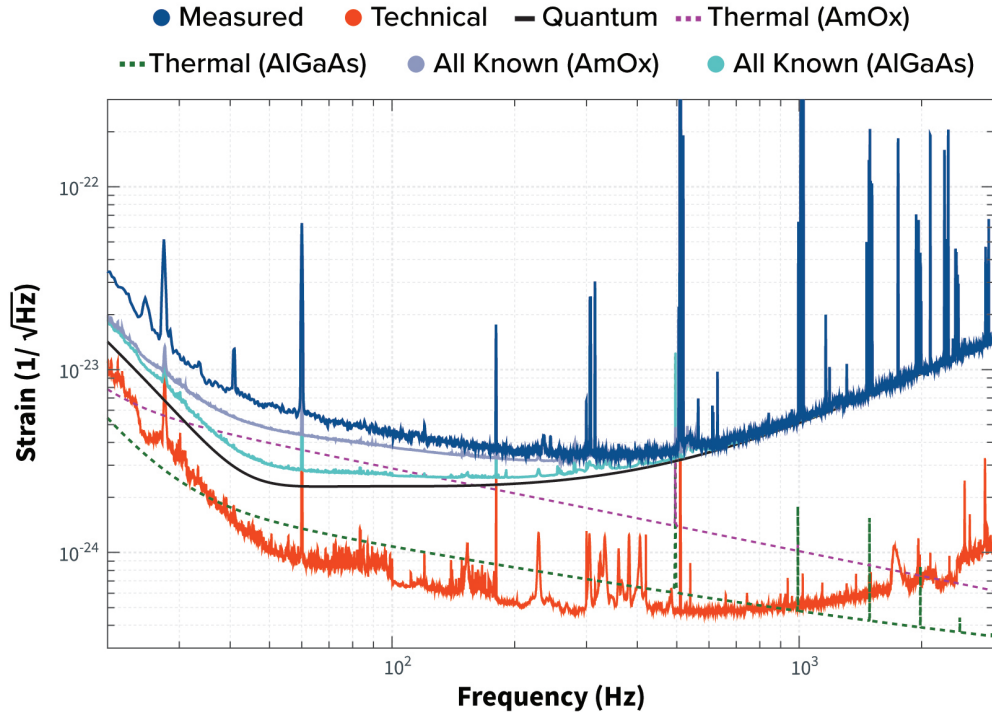


Fig. 7. Noise performance of the Advanced LIGO interferometer in Livingston, LA, adapted from [108]. The Advanced LIGO interferometers use amorphous oxide (AmOx) coatings ($\text{TiO}_2\text{:Ta}_2\text{O}_5/\text{SiO}_2$) but we also show the performance expected if AlGaAs coatings were used instead. The difference between the solid gray and solid teal curves is therefore entirely due to the improved thermal noise performance of AlGaAs. Note that the thermal noise curves here are the combined thermal noise from all sources, including coatings, substrates, suspensions, TO noise, etc. The narrow lines in the thermal noise spectra are due to resonances of the test masses and the silica wires supporting the test masses. The discrepancy between the expected noise (solid, light blue curve) and the measured noise (solid, gray curve) is not yet fully understood but may be partly due to excess CTN. The data for the noise spectra in [108] were kindly provided by Elenna Capote of Caltech/LIGO Hanford Observatory.

439 As a result, research has focused on identifying an alternative high-index material with low elastic
440 loss and low absorption.

441 Another key empirical observation is that room temperature elastic loss generally decreases
442 with increasing post-deposition annealing temperature, until reaching a limit set by the onset of
443 crystallization and/or structural issues such as [delamination](#). As a result, one strategy for reducing
444 elastic loss in amorphous systems has been investigating alloys or mixtures (colloquially referred
445 to here as "dopants"), deposition conditions, as well as nanoscale coating layers (nanolaminates)
446 that may raise the maximum anneal temperature. Experiments have shown that the addition of
447 different dopants to Ta₂O₅ films with a cation content of ~ 25% reduces elastic loss and increases
448 the crystallization temperature [15, 116]. The addition of titania, results in an amorphous oxide
449 mixture with one of the lowest elastic losses plus an increased crystallization temperature.

450 A key observation, obtained by correlating measured elastic losses with the characterization of
451 atomic structures via X-ray scattering, is that the elastic loss is minimized with decreasing fraction
452 of face- and edge-sharing vs. corner sharing polyhedral motifs [120]. Annealing promotes cation
453 diffusion in titania-doped tantalum [17], and hence enables re-arrangements of the polyhedral
454 motifs in the amorphous oxides. These efforts led to the identification of 44%TiO₂:GeO₂ as
455 a promising alternative high-index amorphous coating material for future implementation in
456 GWDs. This is covered in further detail in the Supplementary Document.

457 There remain significant challenges to consistently realize high optical quality in 44%TiO₂:GeO₂
458 HR coatings. This includes variations in the onset of crystallization, film absorption, and proba-
459 bility of film failure that are dependent on process conditions and which have proven difficult to
460 identify and mitigate. A practical issue that complicates the reliable fabrication of these IBS
461 coatings is delamination of the coatings from the substrate that sometimes accompanies the
462 annealing process. A complicated dependence on many process parameters has been observed,
463 among which are base pressure in the deposition chamber [121], substrate temperature during
464 deposition, coating thickness, and substrate polishing and pre-annealing. Gains in extending
465 the onset of crystallization beyond 600 °C will enable further reduction in CTN and absorption;
466 however, surprisingly strong variations in crystallization behavior are currently observed. For
467 example, in GeO₂ films deposited under slightly different IBS conditions, crystallization temper-
468 atures have been observed to differ by ~ 150 °C. An important area of ongoing research is to
469 develop an understanding of the mechanisms responsible for these effects along with conditions
470 that enable reliable use of higher annealing temperatures for further reductions in elastic loss.

471 While this review focuses on IBS oxides and crystalline AlGaAs mirrors, it is important to note
472 that there are ongoing efforts devoted to developing amorphous semiconductors as high-index
473 coating layers for GWDs. Several of these, notably SiN_x and amorphous Si (a-Si), have high
474 refractive indices and sufficiently low elastic losses for these applications, but currently suffer
475 from higher than acceptable optical absorption, the reduction of which is the focus of current
476 research efforts [122–127]. Combinations of materials in multi-material coatings, with low
477 absorption (but mechanically lossy) materials in the top layers, and low elastic loss (but optically
478 absorbing) materials in the lower layers of a stack is one approach to take advantage of disparate
479 material properties [128–130].

480 3.2.2. Substrate-transferred AlGaAs coatings for future GWDs

481 AlGaAs-based crystalline coatings are the most mature of the alternatives to amorphous oxide
482 supermirrors; their study attracts an increasing amount of interest in the GWD community and
483 beyond. To meet the specifications for a 4-km GWD, the coating diameter would need to be
484 scaled to 30 cm and the optical losses and laser damage associated with any crystal defects would
485 need to be small. Therefore current research is focused on qualifying the optical properties
486 (absorption, defect density, birefringence, and birefringence variation) and on reducing the cost
487 of scaling to large area. Fortunately, the thermal noise is already very low, so the detector network

488 would immediately benefit from the performance gain of the reduced CTN.

489 Over the last 10 years, large-area crystalline coating prototypes of increasing diameter starting
490 with 50 mm, then 100 mm, and now up to 200 mm have been delivered for analysis by the GWD
491 community [36, 98, 131, 132]. The optical performance of these mirrors remains promising,
492 with absorption measured at < 1 ppm [40] and integrated optical scatter values < 10 ppm [131].
493 As commercial GaAs wafers are currently unavailable at this size scale, alternative approaches
494 such as heteroepitaxial growth on Ge are currently being explored in order to reach a diameter of
495 300 mm. A key focus in those efforts will be to control and minimize any additional optical losses
496 that may result from increased background impurity levels, as well as surface and interfacial
497 roughness.

498 As discussed in the cavity-stabilized laser section, AlGaAs coatings are known to be birefringent.
499 For a single reflection at normal incidence, orthogonal polarizations can accumulate a phase
500 difference of up to 1.1 ± 0.2 milliradians when the polarization axes are aligned with the
501 $(\bar{1}00)$ and $(1\bar{1}0)$ cleavage planes [36]. The uncertainty is largely due to observed birefringence
502 non-uniformity across AlGaAs coatings, at the level of a few tenths of milliradians. The non-
503 uniformity is associated with the bonding process and is not present in as-deposited coatings on
504 the epitaxial growth wafer. The non-uniformity arises because the coating is unable to slip against
505 the substrate during bonding; this frustrates the ability to relax in-plane stress concentrations
506 that accumulate. While the effects of uniform birefringence can be mitigated through methods
507 like rotational anti-alignment of cavity optics, birefringence non-uniformity represents a source
508 of scatter that cannot be mitigated. Fortunately, in LIGO-like detectors, the optical mode loss
509 from non-uniform birefringence is below the level of concern [36]. [Birefringence non-uniformity](#)
510 [contributes significantly less to the optical mode loss than the expected mirror surface figure](#)
511 [deviations, for example](#). Nonetheless, the effects of birefringence and birefringence variation in
512 GWDs are not fully explored, and reductions in both remain an important goal of ongoing work.

513 4. Laser Damage in High-Performance Optical Coatings

514 Optimizing optical coating technologies for future high average or peak power applications
515 requires additional technical issues to be addressed. In GWDs, there is a clear path towards
516 increased intracavity arm powers [133], while stabilized frequency combs entail the injection of
517 high-peak-power modelocked laser pulses into high-finesse cavities [4, 134]. Failure of optics
518 under these conditions is quantified by the laser induced damage threshold (LIDT), where a
519 maximum fluence is determined from probabilistic tests that identify the onset of damage [135].
520 It is important to note that a practical failure mode may not be manifested by physical damage
521 of the components, but rather degradation due to exposure, which is specific to the operational
522 parameters of the laser system, including the photon energy, as well as the average power, pulse
523 energy, duration, and repetition rate. In optical interference coatings where the electric field
524 distribution is highly modulated, damage initiation is governed by the presence of absorbing
525 precursors in conjunction with the local electric field intensity [136]. The strategies to reduce
526 optical losses and survive >1 MW average powers and intensities of 10 kW/cm^2 over $\sim 100 \text{ cm}^2$
527 beams without damage or figure distortion apply here.

528 For high-average-power laser systems, including continuous wave (CW) or high repetition
529 nanosecond lasers, minimizing absorption is of paramount importance. Such absorption sources
530 can be associated with atomic level defects or localized defects from the manufacturing process.
531 Uniform background absorption can result from dangling bonds, or the incorporation of impurities
532 such as hydrogen or transition metals in amorphous oxides, while in crystalline coatings free-
533 carrier absorption typically dominates, driven by unintentional impurities (e.g., background
534 carbon). Local absorbers, for instance particulate inclusions and nodules in amorphous oxides
535 and oval defects in GaAs-based materials, can result both in local thermal distortions of the
536 reflected wavefront and damage initiation sites limiting peak or average power handling.

537 For pulse durations shorter than about 2 ps, damage initiation is governed by the electric
538 field distribution, and the damage initiation threshold is more deterministic [136]. In this case,
539 the value of the optical bandgap of the coating materials is critical in determining its LIDT.
540 Therefore, most interference coatings for high intensity lasers are a combination of wide-bandgap
541 amorphous oxides, primarily HfO_2 and SiO_2 [137]. Extending these systems to higher peak
542 powers and shorter wavelengths are being explored. Frustratingly, significant variation among
543 nominally identical coating stacks are observed. In amorphous materials, this variability has
544 been assigned in part to the presence of defects states within the bandgap that extend the Urbach
545 tail into longer wavelengths [138, 139]. Such defects can introduce linear absorption of the
546 laser light (for ultraviolet laser wavelengths) or reduce the order of multiphoton absorption, thus
547 lowering the laser intensity required to induce breakdown of the material [139]. The impact of
548 damage initiation on the functional performance of the optic can be very different depending
549 on the aperture size. Considering that the size of a damage initiation site is typically on the
550 order of 1-10 μm , depending on the pulse duration, the impact in large aperture laser beams is
551 negligible. However, the stability of the damage site under subsequent laser irradiation, referred
552 to as “damage growth”, assumes greater prominence. Studies to-date suggest that the damage
553 growth threshold can be significantly lower than the LIDT [140, 141]. Additionally, the presence
554 of contaminating particles on the surface of the optic is of critical importance as they can cause
555 field intensification, leading to damage. This is of particular concern with narrower bandgap
556 coating materials [142].

557 4.1. *Dielectric interference coatings for high average and peak power lasers*

558 Metal oxide interference coatings are integral components in the high peak and average power
559 lasers driving developments in directed energy systems [143], inertial confinement fusion efforts
560 [144], and for pushing the frontiers of high-intensity laser science [145]. These coatings enable
561 exceptional control of the mirror spectral response over the range of $200\text{ nm} < \lambda < 2200\text{ nm}$,
562 while realizing ultra-low optical losses. Further, careful design allows for superior polarization
563 and dispersion control. Optimizing these coating technologies for future high average or peak
564 power applications requires additional technical issues to be addressed as laser damage and
565 degradation during extended exposure is a major limitation for the most advanced laser systems
566 producing pulses from tens of ns to a few fs.

567 4.2. *Laser damage in crystalline coatings*

568 The study of laser damage processes in crystalline coatings is much less mature than that of their
569 amorphous counterparts. Coupled with their very low optical absorption, the increased thermal
570 conductivity of GaAs/AlGaAs interference coatings ($>20\text{ Wm}^{-1}\text{K}^{-1}$) makes them promising for
571 high-average-power CW or quasi-CW laser systems [146]. Operating well below the damage
572 threshold, the ability to more efficiently dissipate absorbed power is beneficial in maintaining a
573 desired surface figure and reducing thermal distortions [147]. Ultimately, under these conditions,
574 the damage threshold is directly related to the ability to extract deposited heat. Using a coating
575 material that offers increased heat conduction is of fundamental importance, but this must also
576 be coupled with high conductivity substrates and with proper thermal management, via the
577 incorporation of passive or active cooling as integral parts of the overall optical assembly.

578 Given the modest bandgap of GaAs (1.42 eV), these optics may be more sensitive to surface
579 contaminants, while pulsed LIDT is limited by multiphoton absorption to $<10\text{ J/cm}^2$ (for ns
580 pulses at a center wavelength of 1064 nm) [146]. Pushing to more robust fieldable optics,
581 as well as higher pulsed LIDT fluences in the NIR and shorter wavelengths will require the
582 implementation of wider bandgap epitaxial systems including the III-nitrides [148] or oxide
583 systems such as Ga_2O_3 . However, current GaAs/AlGaAs-based crystalline coatings should excel
584 for MIR applications. Here, two-photon absorption is eliminated at wavelengths longer than

about 1740 nm, while maintaining ppm-level absorption for wavelengths > 5000 nm [5, 12]. As will be discussed in the following sections, this makes GaAs/AlGaAs crystalline coatings extremely promising for CO₂ lasers at 10.6 μm , where low-optical-loss coating materials are in short supply.

5. Extending Supermirror Technology to MIR Wavelengths

With long-wavelength illumination, light interacts with vibrational modes in the coating materials via phonon absorption, ultimately limiting the optical performance of potential supermirror candidates in this spectral range. For amorphous materials, the lack of long-range order gives rise to broad vibrational absorption bands associated with bond stretching and bending modes. For example SiO₂ exhibits a broad phonon absorption peak around 9800 nm, with a tail extending throughout the MIR [149]. The low atomic mass of oxygen leads to higher vibrational frequencies for O-based bonds compared to those involving heavier atoms such as F, S, Se, or Te as in fluorides and chalcogenides. Fluoride and chalcogenide glasses—including arsenic trisulfide and various selenides—exhibit significantly lower phonon energies than their oxide counterparts, providing better intrinsic transparency for MIR applications and making them well suited as substrate materials. Integration of these materials in high-performance multilayer mirror coatings remains difficult as they are prone to crystallization during thin film deposition.

Significant progress has been made in enhancing amorphous coatings for the MIR, with commercial HR coatings now achieving total losses of $1 - R$ below 30 ppm. For example, $\mathcal{F} \approx 114\,000$ at a wavelength of 4500 nm has been reported in Ref. [150]. While the specific material composition is not quoted in that work, likely options are Si, Ge, ZnSe, ZnS, and various fluorides. Studies such as Refs. [151, 152] provide valuable insights into the suitability of different materials and deposition techniques for this spectral range. With relaxed absorption specifications, IBS optical interference filters [153] and partial [154] or chromatic beam splitters [155] are key components for frequency comb based spectroscopy setups at wavelengths around 8000 nm, given the ability to carefully tailor the coating dispersion.

5.1. Crystalline MIR supermirrors

Investigations of the MIR optical properties of GaAs/AlGaAs crystalline coatings have confirmed exceptionally low optical absorption for these mirrors, with measured values below 10 ppm [5]. This has enabled new scientific discoveries in chemical kinetics via time-resolved frequency comb spectroscopy [156]. As opposed to NIR mirrors using fused silica substrates, MIR GaAs/AlGaAs multilayers are transferred to relevant substrate materials including c-Si, ZnSe, and Ge, depending on the target CWL of the mirrors. Due to their modest refractive index contrast, tens of layer pairs are required to reach a reflectivity well above 99.99% in this system. Moreover, due to the long CWL, the required quarter-wave layer thicknesses lead to a total stack thickness of many tens of microns. As epitaxially-grown crystalline coatings are subject to thickness limitations owing to strain buildup due to the slight lattice mismatch between the low-index AlGaAs and high-index GaAs films, high-quality epitaxial multilayers are limited to a total thickness of $\sim 10\text{--}15\,\mu\text{m}$. For the sake of comparison, a GaAs/AlGaAs quarter-wave DBR designed for a CWL of 4500 nm and a target transmission of 5 ppm requires a total thickness exceeding 30 μm . This limitation can be bypassed by growing half the DBR, then bonding the same stack on top of itself to produce the crystalline coating before transfer to a c-Si optical substrate [5, 12]. In a pioneering experimental effort, two crystalline half-stacks (22.25 periods) were directly bonded to realize a DBR with 44.5 periods of GaAs/Al_{0.92}Ga_{0.08}As with a nominal transmission of $T = 9.3$ ppm and a measured excess loss of $A + S = 4.3$ ppm at a center wavelength 4450 nm [12]. These "all crystalline" coatings ultimately yielded a maximum cavity finesse of $\mathcal{F} = 231\,000$, setting a new performance benchmark for MIR mirrors and enabling significant advancements in cavity-based trace gas detection as discussed in Section 5.4.

633 5.2. Hybrid IBS/crystalline coatings for MIR supermirrors

634 Although successful in enabling ultralow-loss MIR coatings, double-bonded all-crystalline
635 mirrors are limited in terms of uniformity and production yield. This motivated the development
636 of a "hybrid" coating approach that combines the advantages of amorphous multilayers with
637 those of crystalline epitaxial films in one device. These multi-material structures include a
638 high-index-contrast base a-Si/SiO₂ multilayer with a wide stopband capable of high reflectance
639 with only a few layer pairs and a thickness < 5 μm. This amorphous partial mirror is then capped
640 with a direct-bonded ultralow-absorption crystalline coating. This combination of amorphous
641 and epitaxial layers yields a MIR supermirror with ultralow excess loss and high reflectivity,
642 while simultaneously improving uniformity and production yield. As the electric field intensity
643 rapidly decays in a DBR, the higher absorption of the amorphous base stack (with ≈ 1000×
644 larger extinction coefficient) plays a minor role as the field intensity is attenuated significantly
645 after propagating through 22 pairs of GaAs/Al_{0.92}Ga_{0.08}As.

646 Although the impact of surface roughness on scattering is lower at long wavelengths, for
647 hybrid mirror fabrication, successful direct bonding requires excellent surface quality, demanding
648 an RMS micro-roughness < 1 nm to produce a robust amorphous-crystalline interface. Such
649 high-surface-quality base coatings have been realized with super-polished c-Si substrates and
650 dense IBS-deposited a-Si and SiO₂ multilayers [12, 13]. In these experiments, the base IBS
651 coatings exhibit a microroughness of ≤ 1 Å [13]. To date, several material combinations have
652 been investigated for the amorphous sub-stack in MIR hybrid mirror coatings, with a-Si employed
653 as the high-index material and SiO₂ as well as Ta₂O₅ as low-index layers. For the terminating
654 amorphous material (at the bond interface) both a-Si and Al₂O₃ have been used. Among these,
655 the tri-material combination of Ta₂O₅, SiO₂, and a-Si has demonstrated promising performance
656 for AR coatings, as reported in [13].

657 The performance of selected amorphous sub-stack configurations has been quantitatively
658 assessed through cavity ring-down measurements after bonding a crystalline multilayer from
659 the same epi-run. In the configuration using amorphous a-Si and SiO₂ with an Al₂O₃ bonding
660 layer, reported in [12], the total scattering and absorption losses ($S + A = L$) were measured
661 at 5.17 ppm, with a transmission loss of 2.53 ppm. This configuration achieved a two-mirror
662 cavity finesse of 409 000, corresponding to a total per-mirror loss of $1 - R = 7.7$ ppm. In
663 comparison, the configuration employing a-Si and Ta₂O₅ with an a-Si bonding layer, as presented
664 in [13], exhibited slightly higher excess losses of 6.8 ppm, with a similar transmission loss of
665 $T = 2.5$ ppm. The resulting cavity finesse was measured to be 304 000 (total per-mirror loss of
666 9.3 ppm). While SiO₂ exhibits increased absorption at wavelengths above 4.8 μm, Ta₂O₅ appears
667 to remain suitable for the realization of hybrid mirrors operating at even longer wavelengths.

668 5.3. Prospects for wavelengths beyond 5000 nm

669 Pushing deeper into the IR spectral range amplifies existing challenges in mirror design and
670 manufacturing. A key issue involves the limited availability of reliable refractive index data,
671 recently addressed for GaAs and AlGaAs [157], but still unresolved for many other materials. As
672 quarter-wave stacks grow increasingly thick at longer wavelengths, hybrid coatings become the
673 only viable option for high-performance supermirrors, leveraging the low excess losses of the
674 GaAs/AlGaAs crystalline material system as the surface layers and amorphous base coatings in a
675 multi-material stack. With appropriate material selection and optimized manufacturing processes,
676 this platform enables ppm-level excess losses over the entirety of the MIR and extending to
677 CWLs beyond 10 μm. The long-wavelength performance of these coatings is ultimately limited
678 by free-carrier absorption, which is driven by unintentional background doping [40]. While
679 absorption in the sub-coating is less critical, fabrication of the amorphous base stacks remains
680 challenging due to the need for exceptional surface quality for direct bonding [13].

681 For mirror CWLs beyond 5 μm, alternatives to conventional metal oxides and a-Si for the

base coating are being explored, with promising candidates including Ge for the high-index layers and various fluorides, sulfides, and selenides for the low-index layers. As can be seen in Fig. 8, crystalline mirrors have the potential to maintain < 10 ppm of absorption out to a CWL of 6000 nm and < 30 ppm to 8000 nm, enabling cavity finesse values $\mathcal{F} = 100\,000$ over this key spectral range. At $10.6\,\mu\text{m}$, the primary emission line of CO_2 gas lasers, conventional mirror reflectivities currently reach a maximum value of approximately 99.8% (limited by absorption at ~ 2000 ppm). In contrast, GaAs/AlGaAs crystalline coatings promise significant performance enhancements, with an absorption level < 100 ppm, yielding $R > 99.99\%$ and the potential for enhanced LIDT at this wavelength. Such ultralow-loss and high-power-handling long-wave-IR mirrors will have a wide-ranging impact on diverse applications including sensing, manufacturing, materials processing, medical treatments, and the driving CO_2 laser systems employed in extreme-ultraviolet lithography systems.

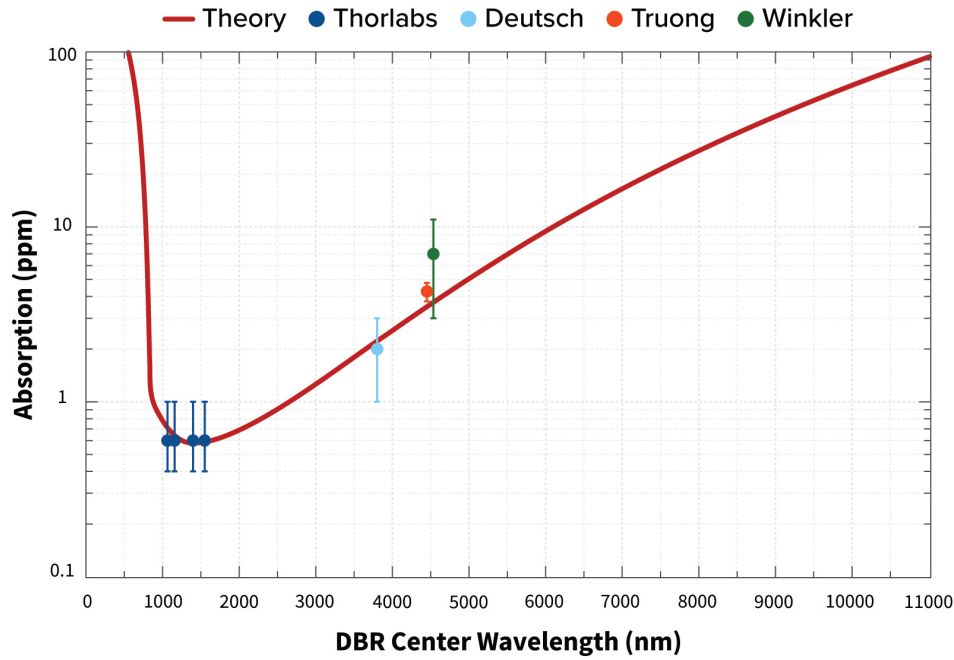


Fig. 8. Semi-logarithmic plot of optical absorption as a function of DBR CWL for MBE-grown unintentionally doped GaAs/Al_{0.92}Ga_{0.08}As mirrors from 500 nm to $10.6\,\mu\text{m}$ at room temperature. The theoretical curve (red) incorporates the wavelength-dependent linear absorption coefficient, the refractive index dispersion of the high- and low-index layers, and the corresponding variation in mirror penetration depth [2]. The material absorption is averaged over the layer pairs and is based on a phenomenological model incorporating two components: i) an Urbach-tail term describing the exponential roll-off near the GaAs band edge (~ 870 nm), and ii) a free-carrier absorption term assuming a conservative λ^3 dependence for modestly doped material where ionized-impurity scattering dominates. Data points include: dark blue – typical absorption values for Thorlabs catalog semiconductor supermirrors with CWLs of 1064 nm, 1156 nm, 1397 nm, and 1550 nm [158]; light blue – photothermal common path interferometry measurement of a 3800 nm transferred AlGaAs DBR From Deutsch et al. [159]; orange – the all-crystalline mirror reported in Truong et al. [12]; and green – results from Winkler et al. [5].

694 5.4. High-sensitivity trace gas detection in the fingerprint region

695 Optical trace-gas sensing has seen a dramatic improvement in sensitivity with the advent of
 696 cavity-enhanced techniques such as cavity ring-down spectroscopy (CRDS) [160] and noise-
 697 immune cavity-enhanced optical-heterodyne molecular spectroscopy (NICE-OHMS) [161]. For
 698 early books on the subject, see References [162, 163]. These linear absorption spectroscopy
 699 methods leverage an increased light-matter interaction path length and importantly decouple
 700 measurements from laser amplitude fluctuations by either recording the passive exponential
 701 decay of light leaking from the cavity (CRDS) or by using frequency modulation techniques to
 702 avoid fluctuations (NICE-OHMS). In both cases, the enhancement cavity serves to extend the
 703 interaction path length by several orders of magnitude, significantly improving overall sensitivity
 704 for trace gas detection and other applications [164]. The path-length enhancement factor, $\mathcal{E}_{\text{path}}$,
 705 for transmission of a laser tightly locked to a two-mirror cavity with finesse, $\mathcal{F}$, is given by:

$$\mathcal{E}_{\text{path}} = 2 \frac{\mathcal{F}}{\pi} = \frac{2}{T + L}, \quad (4)$$

706 For example, an absorption path length of nearly 100 km can be achieved in a table-top
 707 instrument by using a cavity of 1 m in length and with $\mathcal{F} \approx 150\,000$. Note that, for CRDS, $\mathcal{E}_{\text{path}}$
 708 is lower by a factor of two [165].

709 The above-mentioned linear absorption spectroscopy methods can be extended to multi-species
 710 detection over broader spectral bandwidths via the coherent coupling of optical frequency-comb
 711 laser sources to enhancement cavities. Several variations of CRDS have been performed with
 712 optical frequency combs [166–170] and noise-immune cavity-enhanced optical frequency comb
 713 spectroscopy [171, 172] has also been demonstrated, showing that a combination of high
 714 sensitivity and broad spectral coverage is enabled by the use of high-finesse optical resonators
 715 and low-loss mirrors. The coupling of frequency combs and cavities also offers intriguing
 716 possibilities for ultrasensitive transient absorption spectroscopy [173] and multidimensional
 717 spectroscopy [174], with additional finesse scaling advantages for both the pump and probe
 718 beams which are not covered here.

719 Nonlinear absorption, frequency-domain spectroscopy techniques that also leverage cavity
 720 enhancement have demonstrated further improvements in sensitivity for trace gas detection, in
 721 some instances reaching parts-per-quadrillion (ppq) levels. These approaches, like saturation
 722 absorption cavity ring-down spectroscopy (SCAR) [175, 176], two-color CRDS [177, 178],
 723 and near-resonant two-photon CRDS [179], not only make use of the path length enhancement
 724 factor but also exploit the enhancement of the average circulating power within the high-finesse
 725 resonator and sample cell. This complimentary enhancement enables isotope-selective detection
 726 at levels that rival state-of-the-art mass spectrometry. For a tightly locked cavity, the power
 727 enhancement factor, $\mathcal{E}_{\text{power}}$, is given by [4, 180]:

$$\mathcal{E}_{\text{power}} = \frac{T\mathcal{F}^2}{\pi^2} = \frac{T}{(T + L)^2}. \quad (5)$$

728 Together, the enhancement factors $\mathcal{E}_{\text{path}}$ and $\mathcal{E}_{\text{power}}$ highlight the mirror characteristics most
 729 useful for designing cavity-enhanced laser spectrometers and sensors for trace gas detection: low
 730 total losses, $T + L$, and low excess losses, L . Note that the optical power transmitted through
 731 the cavity in these on-resonance, laser-locked examples is simply $T\mathcal{E}_{\text{power}}$, and its magnitude
 732 will influence the achievable measurement signal-to-noise ratio (e.g., [181, 182]), and therefore
 733 sensitivity, of both linear- and nonlinear absorption methods.

734 Cavity-enhanced spectroscopy and sensing techniques critically depend upon the availability
 735 of low-loss, high-reflectivity mirrors to achieve long effective path lengths and high intracavity
 736 powers. In the MIR, realizing low-loss optics with cavity finesse values exceeding several
 737 thousand had long been a significant challenge, with amorphous materials being limited by

intrinsic optical absorption, while additional losses stemmed from surface-adsorbed water in hydrophilic coatings [183], or from water absorbed in the surface layer due to increased porosity in under-dense layer materials [184]. These limitations posed particular difficulties in studies involving gas mixtures that contained both strongly absorbing species with sharp spectral features and components with broader, less distinct signatures, which were often buried beneath the noise floor (e.g., [185]). To illustrate the impact of excess losses, simulations of theoretical $\mathcal{E}_{\text{path}}$ and $\mathcal{E}_{\text{power}}$ values are plotted in Fig. 9 for a hypothetical two-mirror cavity with fixed $T = 25$ ppm, along with selected reference points taken from the literature. Compared with traditional PVD-derived amorphous coatings, epitaxial coatings exhibit reduced bulk optical absorption and enhanced surface stability (i.e., lack of porosity and thus hygroscopicity) due to the fully densified nature of a crystalline solid.

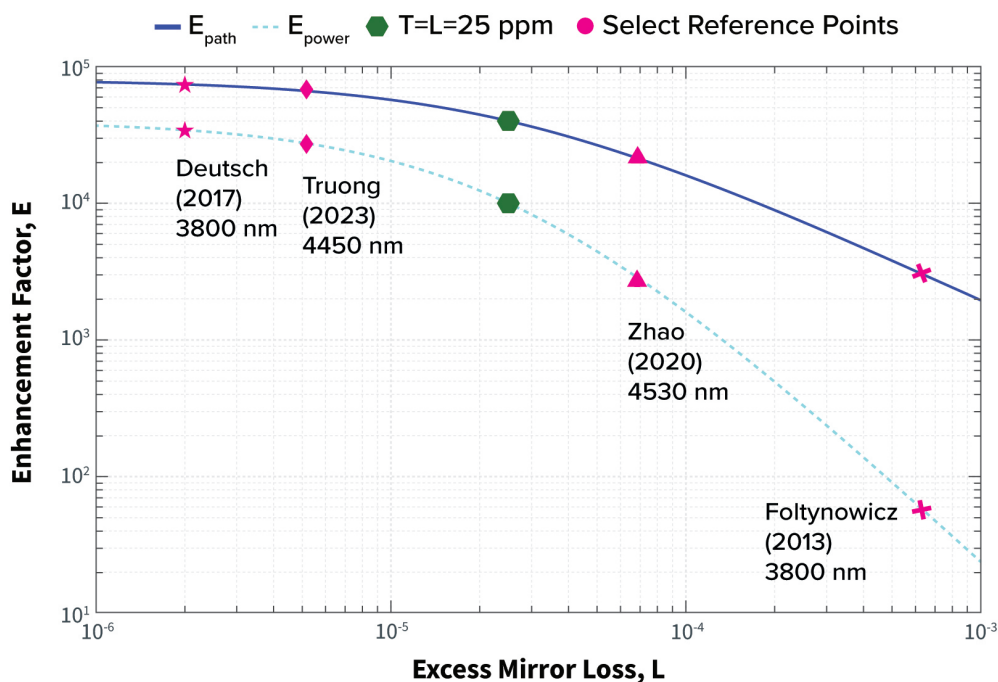


Fig. 9. Illustration of theoretical enhancement factors, $\mathcal{E}_{\text{path}}$ and $\mathcal{E}_{\text{power}}$, for a hypothetical two-mirror cavity with fixed mirror power transmission coefficient $T = 25$ ppm. Also indicated is the case where the excess power loss coefficient, L , is equal to T , as well as several selected reference points taken from the experimental literature where T and L have been measured.

A key experiment that demonstrated the potential of low-loss MIR optics was the direct frequency comb investigation of the $\text{OD} + \text{CO} \rightarrow \text{DOCO}$ reaction kinetics and branching ratios [156, 186]. This work not only advanced the field of MIR spectroscopy but also marked a breakthrough for crystalline coating technology in this spectral region. Furthermore, the development of low-loss coatings—particularly the hybrid structures discussed in Section 5.3—is now enabling a new generation of high-sensitivity, high-precision applications of spectroscopy in the MIR.

For example, critical line lists comprising spectral reference data enable consistency in measurements across diverse systems like dynamic atmospheres, astrophysical objects, high-purity industrial gases, and extreme combustion and plasma environments. Precision molecular

759 spectroscopy in the MIR, enabled by low-loss mirror coatings—is now poised to test line lists
 760 from phenomenological, semi-empirical, or *ab initio* quantum chemistry calculations in new
 761 regimes. Recently, low-loss mirror coatings were applied in CRDS measurements of a carbon
 762 monoxide (CO) transition intensity at a wavelength near 4530 nm [187]. An example spectrum
 763 from the recent work is reproduced here in Fig. 10.

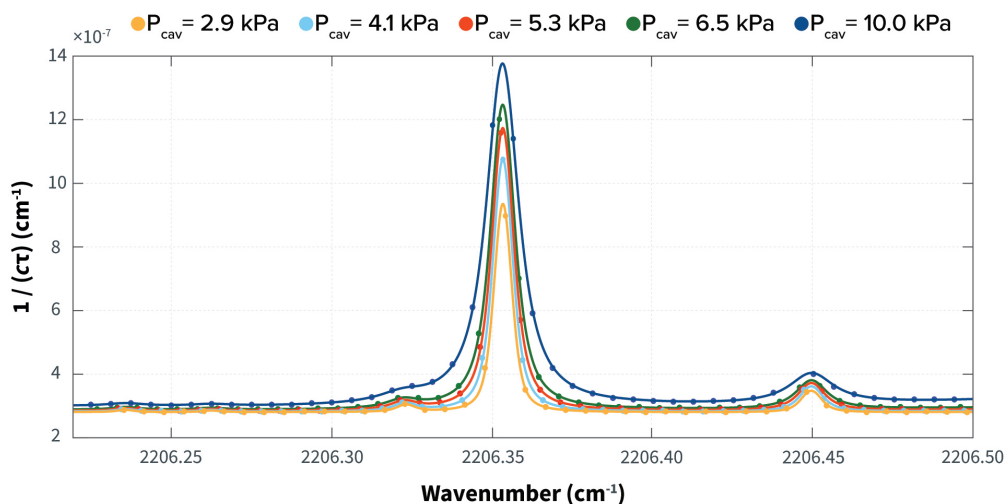


Fig. 10. The rotationally resolved cavity ring-down spectrum of carbon monoxide (CO) fundamental vibrational band near a wavelength of 4530 nm. Also visible are weakly absorbing features from nitrous oxide (N₂O) present in the air sample. Figure is reproduced from data reported in Bailey et al. [187].

764 Molecular transition intensities are quantities proportional to the Einstein coefficient for
 765 spontaneous emission (Einstein-A coefficient) of a particular rotational-vibrational transition,
 766 and therefore proportional to a fundamental property of the molecule being probed by light.
 767 Performed using a certified gas sample of CO-in-air, the recent study by Bailey et al. reported a
 768 new value for the CO transition intensity with sub-percent relative standard uncertainty [187].
 769 Importantly, the chosen rotational-vibrational transition was within the fundamental vibrational
 770 band of CO, a band from which spectral reference data is now routinely relied upon to analyze
 771 data from IR astronomical observatories such as the James Webb Space Telescope [188–190].
 772 The high-fidelity and traceability of CRDS measurements in the MIR revealed discrepancies
 773 between the prior works used to scale popular line lists [191–193], and motivated ongoing efforts
 774 to improve the quality of the low-uncertainty data used broadly by the sensing and observational
 775 communities [194].

776 Taking an educated glimpse into the future, we anticipate several avenues of progress in
 777 high-sensitivity trace gas detection within the fingerprint region. Mirror, laser, and detector
 778 technologies are expected to mature further, extending reliably into the 5–10 μm range, where
 779 molecular absorption features are typically stronger and more distinct than in the 3–5 μm region.
 780 This shift will enable enhanced sensitivity and selectivity in spectroscopic measurements. We
 781 expect all established high-sensitivity techniques to continue advancing into this spectral domain.

782 In parallel, novel methods—enabled by low-loss, high-finesse mirrors and recent improvements
 783 in MIR light sources—may open the door to broadband, comb-mode-resolved saturation
 784 spectroscopy. This approach holds the potential to significantly expand the availability of
 785 high-resolution spectral reference data [134].

786 6. Concluding Remarks

787 In conclusion, advancements in high-performance amorphous and monocrystalline optical interfer-
788 ence coatings have fundamentally redefined the limits of precision interferometry and spectroscopy.
789 As reviewed here, ion-beam sputtered amorphous oxide coatings now routinely achieve ppm-level
790 optical losses with exceptional uniformity at large scale, while substrate-transferred crystalline
791 multilayers introduce a qualitatively new capability: ultralow mechanical dissipation combined
792 with comparably low optical loss. Together, these technologies underpin the critical optical
793 resonators used in modern laser-based metrological systems, driving significant gains in the
794 stability of optical atomic clocks, the sensitivity of the interferometric GWDs, and the detection
795 limits of cavity ring-down spectrometers.

796 In the near infrared, optical losses including absorption and scatter approach their practical floor,
797 shifting performance limits toward thermomechanical fluctuations. This transition has catalyzed
798 the adoption of crystalline coatings in ultrastable reference cavities, yielding order-of-magnitude
799 reductions in coating elastic loss pushing thermal noise and fractional frequency instabilities
800 to the low 10^{-17} level in cryogenic systems. In GWDs, coating Brownian noise remains a
801 dominant limitation, making both next-generation amorphous oxides and scalable crystalline
802 mirrors critical for future sensitivity upgrades.

803 Equally transformative is the extension of supermirror technology into the MIR spectral region.
804 Hybrid crystalline/amorphous coating architectures now deliver ppm-level losses across the
805 molecular fingerprint region, enabling unprecedented path-length and power enhancement for
806 high-precision spectroscopy and trace-gas sensing. Looking forward, continued progress will
807 hinge on advances in materials synthesis, large-area manufacturing, and hybrid integration,
808 positioning optical coatings as a central enabling technology for the next generation of precision
809 optical science.

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839 **7. Si: Details of the substrate-transfer crystalline coating process**

840 For the production of such “semiconductor supermirrors” [195], epitaxial growth via molecular beam
841 epitaxy (MBE) is preferred for its lower unwanted impurity level, which is beneficial in reducing free-carrier-
842 mediated optical absorption. Vapor-phase epitaxial growth processes, notably organometallic vapor-phase
843 epitaxy (OMVPE), also known as metal-organic chemical vapor deposition (MOCVD), have also been
844 explored, with promising results, but the lowest absorption values attained remain at the 10 ppm level [196].
845 The background carrier concentration is ultimately lower via MBE; however, a well-known drawback of
846 this process is the formation of oval defects in GaAs growth, arising from spitting of the gallium effusion
847 source [197, 198]. The density of such defects can be significantly reduced through process optimization
848 and careful choice of crucible materials [199, 200].

849 Following epitaxial growth, the substrate-transfer process entails cleaning and inspection of base substrate
850 and multilayer using standard solvents, detergents, and deionized water, processes akin to those used for
851 pre-PVD coating. Prior to bonding, a plasma activation process [201] is carried out on both the epi surface
852 and base substrate. These hydrophilic treated surfaces are then pressed into contact forming an initial van
853 der Waals bond. Bonding can be carried out in ambient or in vacuum, typically at the 10^{-3} mbar range,
854 though processes can also be carried out down to 10^{-6} mbar; contact in vacuum can aid in minimizing
855 interfacial void formation. The bond is strengthened by an annealing process. The plasma activation process
856 significantly relaxes the temperature requirements for the anneal, enabling a high bond strength, quantified
857 by interfacial energies ≥ 1 J/cm² in select material combinations for anneal temperatures < 150 °C . A
858 detailed overview of wafer bonding processes can be found in Refs. [202, 203]. The final process step
859 involves selective chemical etching to remove the remnant GaAs wafer and embedded high-Al content
860 AlGaAs etch stop layer. The extreme selectivity of HF etching of AlGaAs over GaAs [204], results in the
861 recovery of a 0.1 nm RMS microroughness surface.

862 **8. Si: Optimization of amorphous coatings via atomistic modeling and experi-** 863 **ments**

864 Noting that the low-loss material a-SiO₂ is strongly corner sharing, this observation led to the investigation
865 of a-GeO₂ (isostructural to a-SiO₂) as a promising low-loss and high refractive index option [205]. After
866 experimental verification, doping studies were carried out to further raise the refractive index to a level
867 practical for use in a DBR [111, 205]. Careful characterization of GeO₂ films with various alloy fractions
868 of Ti, including measurements of the refractive index as well as the frequency dependence of the loss angles
869 for bulk and shear moduli, led to the identification of 44%Ti atomic ratio as the optimum composition. The
870 Brownian CTN predicted with these optimized properties met the goal of a two-fold reduction planned for
871 the next upgrade of the LIGO detectors [111]. This composition, 44%Ti:GeO₂, annealed at 600 °C for
872 100 hours, achieved the lowest value of average elastic loss angle among high-index amorphous oxide alloys
873 reported to-date, 0.98×10^{-4} [111].

874 Atomistic models and experiments have shown that Ti doping does not functionally compromise the
875 corner-shared network organization of a-Ge-O₂ and that even as deposited 44%TiO₂:GeO₂ exhibits an
876 organization containing predominantly large (>4) rings of corner-shared tetrahedral GeO-Ti connections;
877 further studies of the evolution of structure with annealing are ongoing [206]. Dielectric coating stacks of
878 44%TiO₂:GeO₂/SiO₂ designed to meet the specifications of the end-test mass mirrors have been fabricated
879 [121]. The Colorado State University group has transitioned the details of the deposition process to
880 the Laboratoire des Matériaux Avancés (LMA) who will be coating the test masses for aLIGO+. IBS
881 TiO₂:GeO₂/SiO₂ HR coatings produced by LMA have reached an absorption loss of 0.5 ppm at $\lambda = 1064$ nm,
882 scatter loss of 30 ppm and measured CTN of 59% that of the TiO₂:Ta₂O₅ A-LIGO mirrors.

883 The insight atomistic models provide on the atomic network organization of sputtered amorphous thin film
884 oxides coupled with control in the synthesis and comprehensive diagnostics are guiding next steps towards
885 developing thin films with further reductions in elastic loss and absorption. Beyond ultra-stable cavity
886 applications, such advances will have impact across the field of high-performance dielectric amorphous
887 oxides, prevalent in the most demanding laser science applications.

9. SI: Mirror coatings for third generation GWDs

Third generation GWDs such as Cosmic Explorer (CE) and the Einstein Telescope (ET) require larger and heavier optics up to 70 cm in diameter, 30-40 cm thick, weighing hundreds of kilograms. Benches working at 1064 nm compatible with these larger optics are already available at LMA including a scatterometer, an absorption test station (employing photothermal deflection), surface defect measurement, and an interferometer. These instruments can be adapted easily if a new reference wavelength is used in the future (1.5 μm for example). A new IBS coating machine will be available at LMA in 2028, which will be able to produce high uniformity amorphous coatings simultaneously on two substrates of 80 cm diameter (load capacity > 500 kg).

CE with arm lengths of 40 km, and increased stable radius of the optical cavity mode will require coatings with thermal noise of second generation detectors but with strict optical absorption requirements (≤ 0.1 ppm) to keep thermal figure distortions manageable in the presence of the high optical powers (1.5 MW). The mirrors also must survive average intracavity intensities $\sim 10 \text{ kW cm}^{-2}$ without damage to support these arm powers. Determining the origin of optical absorption at the sub-ppm level in mirrors and the statistics of local absorbing centers, and developing means to reduce them is therefore a focus of current research.

A CE with 20-km arm length would relax the size requirement (~ 50 cm diameter), but would require a reduction in thermal noise by a further factor of two beyond that of the second-generation mirrors to reach design sensitivity. A similar reduction in thermal noise would be important for any detectors installed in the existing 4-km infrastructure, such as the proposed A#-upgrade for LIGO [207], LIGO Arundha (India) [208], the high-frequency Einstein telescope detector [209, 210]. Cryogenic detectors, including KAGRA (Japan) [211], or the low-frequency ET detector, require mirrors with low thermal noise at low temperatures, a particular challenge for amorphous materials, whose elastic loss typically peaks at temperatures $\lesssim 100$ K.

The future GWD network will consist of long (>10 km) and short (<10 km) detectors, operating at both room and cryogenic temperatures. It is therefore unlikely that a single coating technology can satisfy the performance requirements of all the detector types. IBS deposited amorphous oxides appear promising for room temperature operation of the longest interferometers with very large test masses, while substrate-transferred crystalline coatings appear promising for shorter detectors with stricter CTN requirements. As discussed previously, hybrid coatings that combine disparate technologies are also likely to play a role.

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