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Hydrogen-induced ultra-low optical absorption and mechanical loss in amorphous silicon for gravitational-wave detectors

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The sensitivity of gravitational-wave detectors is limited by the mechanical loss associated with the amorphous coatings of the detectors' mirrors. Amorphous silicon has higher refraction index and lower mechanical loss than current high index coatings, but its optical absorption at the wavelength used for the detectors is at present large. The addition of hydrogen to the amorphous silicon network reduces both optical absorption and mechanical loss for films prepared under a range of conditions at all measured wavelengths and temperatures, with a particularly large effect on films grown at room temperature. The uptake of hydrogen is greatest in the films grown at room temperature, but still below 1.5 at.% H, which show an ultra-low optical absorption (below 10 ppm) measured at 2000 nm for 500-nm-thick films. These results show that hydrogenation is a promising strategy to reduce both optical absorption and mechanical loss in amorphous silicon, and may enable fabrication of mirror coatings for gravitational-wave detectors with improved sensitivity.

Gravitational waves (GW) were first detected by the Laser Interferometer Gravitational-wave Observatory (LIGO) in 2015 [1]. Continuous improvement of the sensitivity of GW detectors has accelerated the discovery of astronomical events [2]. However, between 30 and 500 Hz the sensitivity of the detectors, which are kilometer-scale variants of Michelson interferometers that measure changes in the distance between highly reflective coated mirrors, is limited by thermal noise of the mirrors coatings [3]. The current LIGO and Virgo detectors operate at room-temperature and 1064 nm, and consist of thick fused silica substrates coated with alternating layers of amorphous (*a*-) silica (SiO_2) and titania-doped tantala (TiO_2 -doped Ta_2O_5); low and high refractive index layers, respectively [4, 5]. This configuration provides highly reflective dielectric mirrors with extremely low optical absorption. The dominant mid-band source of noise in this system is thermal noise proportional to the mechanical loss in the coatings [6], which is dominated by the high refractive index layer that limits the resolution of the detectors. To achieve increased sensitivity for future detectors therefore requires improving or replacing the high refractive index layer.

Amorphous silicon (*a*-Si) is a potential candidate to replace the high refractive index layer in future GW detectors; it yields a two-fold reduction in mechanical loss at room temperature (1×10^{-4} [7] vs 2×10^{-4} [8]) and higher refractive index at 1064 nm (3.60 [9] vs 2.08 [10]). The replacement of the high-index layer by *a*-Si implies that fewer pairs of layers, as well as thinner high-index material layers, are needed to achieve the high reflectivity required for GW mirrors [11]. The reduction of the number of layers and their thickness in turn reduces the overall coating thermal noise.

In addition to low mechanical loss, the coatings also require optical absorption of a few parts per million (ppm) to avoid the many deleterious effects at the very high power used for the interferometry [8]. Optical absorption, which depends on the wavelength of the laser, causes steady-state thermal-expansion-driven distortions of the mirror and hinders the interferometers ability to achieve lock. The optical absorption of *a*-Si is considerably higher at 1064 nm than for oxides, specifically $> 2 \times 10^4$ ppm for 500-nm-thick *a*-Si [12] vs < 5 ppm for 4.7- μm -thick *a*- TiO_2 -doped *a*- Ta_2O_5 [13] or < 9 ppm for 200-nm-thick *a*- SiO_2 [14], due to the lower band gap and higher density of electronic defects in *a*-Si, such as dangling bonds, which lead to unpaired electrons. In our previous work [15], an improved design for LIGO Voyager (a cryogenic GW detector that operates at 123 K and 2000 nm wavelength) was proposed, using *a*-Si/*a*- SiO_2 coatings and crystalline Si (*c*-Si) substrates. However, the optical absorption of *a*-Si is at present too large.

It is therefore necessary to find strategies that reduce the optical absorption of *a*-Si and that do not increase its mechanical loss. The different known strategies to approach this problem consist of (i) using multilayer coatings, in which *a*-Si is used as the high index layer in lower layers of the mirror where the optical intensity is significantly reduced [16–19], (ii) adding some N and O to *a*-Si to increase the film band gap [20], and (iii) increasing the wavelength of the laser illuminating the interferometer [21, 22]. In the relevant range of wavelengths (1064 to 2000 nm), absorption has been suggested to correlate with the dangling bond density ρ_{DB} [23]. It is known that hydrogen is relatively easy to introduce into the *a*-Si network, becomes highly mobile at temperatures above ~ 150 °C, and is effective at reducing ρ_{DB} [24].

In this letter, we show the very positive effect of hydrogen on both optical absorption and mechanical loss, and study how growth temperature and subsequent processing (as-deposited, aged, annealed, and hydrogenated) reduce optical absorption and mechanical loss (at cryogenic and room temperatures) of evaporated *a*-Si films. Optical absorption is reduced with these processing steps and with higher laser wavelength, from 1064 to 2000 nm. The hydrogenation of 500-nm-thick *a*-Si films grown at room temperature yields an ultra-low optical absorption (< 10 ppm) at 2000 nm. Additionally, we show that hydrogenation further reduces the films' already low mechanical loss at all temperatures.

Amorphous silicon films were grown by electron-beam evaporation at a base pressure of 10^{-9} Torr, 0.5 Å/s and three different growth temperatures: 50, 225 and 425 °C. Films were characterized in their as-deposited, aged, annealed, and hydrogenated states. We used elastic recoil detection analysis (ERDA) to quantify the hydrogen content in the films, profilometry to measure their thickness, ellipsometry to measure their refractive index n and electron paramagnetic resonance to quantify their dangling bond density ρ_{DB} . Additional details are described in the Supplemental Material [25].

Optical absorption measurements were performed at room temperature using a modified photothermal common-path interferometric (PCI) technique and lasers with wavelengths of 1064, 1550 and 2000 nm [29]. Our results account for the interference effect due to the refractive index difference between film and substrate [25]. This experimental setup yields the films' optical absorption with a sensitivity of ppm level. Considering the contribution to the PCI signal from the substrate, the detection threshold was estimated as 10 ppm at 2000 nm. The absolute error of the measurements that includes all the alignment and calibration uncertainties was $\pm 7\%$. *a*-Si films 500-nm-thick were grown on 1-in-diameter and 1/4-in-thick Corning 7979 high purity fused silica discs.

Mechanical loss Q^{-1} measurements were performed from 1 to 100 K using c-Si microfabricated double-paddle oscillators (DPOs) [30, 31]. This technique uses an anti-symmetric, very-high quality factor pure shear mode of a mechanical resonator to measure the energy loss in a film. The loss is calculated by comparing the resonator ring-down decay time before and after the film is deposited. *a*-Si films 300-nm-thick were deposited on the DPOs sensing area. At room-temperature, Q^{-1} was measured by the gentle nodal suspension method (GeNS) [32], where 2-in-diameter 1-mm-thick silica wafers are measured in vacuum, before and after the deposition of 500-nm-thick *a*-Si films, with their resonant modes excited by an electrostatic actuator from 1 kHz up to about 30 kHz. The decay time of each mode is determined by the energy loss rate, which allows extraction of the film's mechanical loss.

Figure 1(a) shows the optical absorption, measured at 1550 nm, as a function of growth temperature for *a*-Si

films in the different states. As-deposited films show a reduction of the optical absorption from 2×10^4 ppm to 5×10^3 ppm with increased growth temperature for films grown from 50 °C to 425 °C. Aging the films up to 15 months in air at room-temperature reduces the optical absorption by a factor of ~ 2 for all growth temperatures. Annealing the films in high vacuum for 3 hours at 425 °C causes significant further reduction in optical absorption for all growth temperatures, with the largest reduction (a factor of 10) found for films grown at 50 °C; a fact that we attribute to the $\sim 10\%$ lower atomic density of these films compared to those grown at higher temperature [33]. Figure 1(a) also shows the optical absorption, measured by PCI, of 1- μ m-thick *a*-Si films grown by reactive low voltage ion plating (RLVIP), at 250 °C, before and after annealing at 500 °C [22]. The optical absorption of these RLVIP *a*-Si films has also been corrected as described elsewhere [25], which enables a quantitative comparison with the present work. Annealing has a similar effect in both sets of films.

Figure 1(a) also shows that the addition of hydrogen through the hydrogenation process further reduces the optical absorption of all films at all growth temperatures; with a particularly large reduction for the film grown at 50 °C and only a small reduction for the film grown at 425 °C. We note that the sequential processing of the films is not required to achieve the optical absorption achieved in the hydrogenated state shown in Fig. 1(a); hydrogenation from the as-deposited state yields the same results. Figure 1(a) highlights how temperature and hydrogen, separately, reduce the optical absorption of *a*-Si.

After the hydrogenation process, ERDA on films grown at 50 °C show that hydrogen is uniformly distributed with a concentration of 1.45 ± 0.15 at.% H, while films grown at 425 °C have significantly less H (0.55 ± 0.22 at.% H) overall, which concentrates near the film's surface [25]. The higher hydrogen concentration in films grown at lower temperature is likely related to their somewhat lower atomic density, which facilitates the hydrogen uptake process; annealed films grown at 50 °C are $\sim 10\%$ less dense than films grown at 425 °C [33]. Longer hydrogenation times of the films grown at 425 °C, up to 60 hours, did not further reduce their optical absorption nor significantly modify their hydrogen depth profile [25].

The optical absorption at 1550 nm of *a*-Si grown at 50 °C is reduced 37 times after hydrogenation (from 16,500 to 450 ppm), while ρ_{DB} reduces 5 times (from 4×10^{18} to 8×10^{17} spins/cm³). The optical absorption of the *a*-Si films grown at 425 °C is reduced 4 times after hydrogenation (from 3,700 to 1,050 ppm), while ρ_{DB} is reduced 2 times (from 2×10^{18} to 1×10^{18} spins/cm³). Variations in ρ_{DB} between the as-deposited, aged and annealed states are within error bars, despite significant changes in absorption, and only after hydrogenation a significant reduction in both ρ_{DB} and optical absorption is observed.

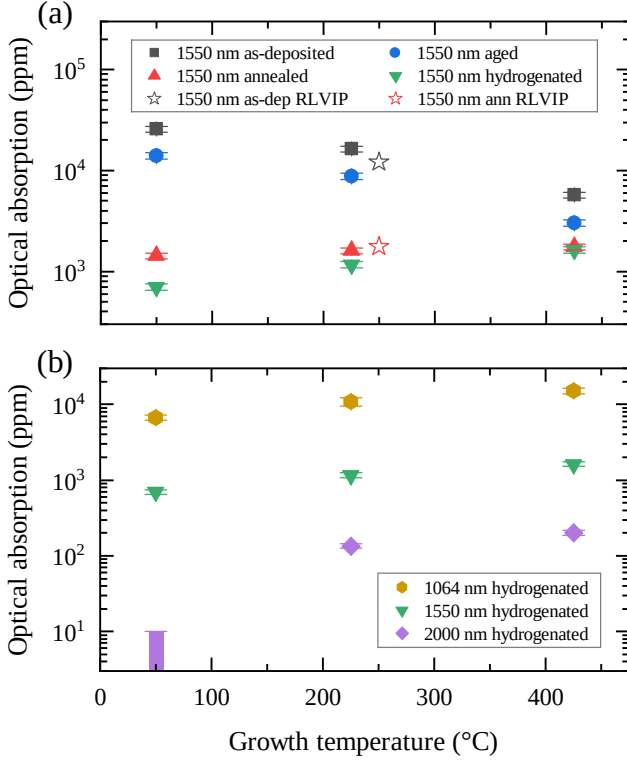


Figure 1. (a) Optical absorption at 1550 nm of 500-nm-thick *a*-Si films in the as-deposited (black squares), aged (blue circles), annealed (red upward-pointing triangles), and hydrogenated (green downward-pointing triangles) states as a function of growth temperature. The optical absorption of 1- μ m-thick *a*-Si films, rescaled to 500 nm (see text for details), grown by RLVP at 250 °C (open star symbols) is also shown in the as-deposited (black) and annealed at 500 °C (red) states [22]. (b) Optical absorption of 500-nm-thick *a*-Si films in the hydrogenated state as a function of growth temperature measured at 1064 nm (yellow hexagons), 1550 nm (green downward-pointing triangles) and 2000 nm (purple diamonds). The purple diamond symbol for the film grown at 50 °C and measured at 2000 nm has been replaced by a rectangle because its value is < 10 ppm.

Figure 1(b) shows the optical absorption of *a*-Si films in their final hydrogenated state measured using wavelengths of 1064, 1550, and 2000 nm. For all wavelengths, optical absorption is lowest in the hydrogenated state, with extremely low absorption (below 10 ppm) measured at 2000 nm for the film grown at 50 °C, which is the lowest optical absorption of *a*-Si reported to date. The substrate contribution of ~ 10 ppm/cm prevents us from quantifying optical absorption below that limit. The optical absorption of *a*-Si films at 1550 nm in the as-deposited, aged, annealed and hydrogenated states, Fig. 1(a), is representative of the trends seen at 1064 and 2000 nm, shown in Fig. 1(b), for the hydrogenated state.

Mechanical loss Q^{-1} of *a*-Si films measured by DPOs, at 1 and 100 K, and by GeNS, at 290 K, is shown in Figure 2 as a function of growth temperature for the as-

deposited, annealed, and hydrogenated states. At 1 K Q^{-1} is due to tunneling states [34, 35], while at 100 and 290 K Q^{-1} is due to thermally-activated states [36, 37]. The effect of growth temperature as well as the evolution of Q^{-1} for each state is shown: at 1 and 100 K, Figs. 2(a) and 2(b), annealing and hydrogenation mildly reduce Q^{-1} (factors of ~ 2), while increasing the growth temperature causes large reductions (orders of magnitude). At 290 K, Fig. 2(c) and its inset, the effect of increased growth temperature is still visible but smaller than at 1 and 100 K; annealing and hydrogenation reduce Q^{-1} (factors of ~ 2) for films grown at 50 and 425 °C. Q^{-1} measured at 290 K does not depend on growth temperature after annealing and hydrogenation, while lower temperature Q^{-1} (1 and 100 K) depends strongly on growth temperature, even after annealing and hydrogenation. The lowest Q^{-1} at each measured temperature is thus obtained for the hydrogenated films regardless of their growth temperature, but the lowest Q^{-1} overall is found for hydrogenated films grown at 425 °C.

We have shown that after annealing and hydrogenation, the optical absorption drops significantly in films grown at 50 and 225 °C, but is reduced only slightly in films grown at 425 °C, such that optical absorption is lower in films grown at lower temperatures. These results correlate strongly with the films' atomic density and H uptake, where the films grown at 425 °C show that little H is incorporated. Results from Raman spectroscopy, summarized in Table I, show that both $\Delta\theta$ and $\text{tr}(\epsilon)$ reduce upon annealing and upon hydrogenation for all growth temperatures, which suggests that temperature and hydrogen, separately, promote the structural relaxation of the *a*-Si films. We note that after hydrogenation, the magnitude of change in Q^{-1} correlates with the H content, which suggests that H atoms play a role in the suppression of the structures responsible for the energy dissipation in *a*-Si.

Early work by Cody et al. [38] studied the optical absorption of hydrogenated amorphous silicon (*a*-Si:H) from 1.4 to 2.0 eV (886 to 620 nm), near the band edge, and suggested that the *a*-Si structure, rather than the at.% H, is responsible for the optical absorption. In this model, the role of H is an indirect one, associated with lowering the Si-Si energy barriers to structural relaxation [39], which in turn modifies the band gap and, therefore, the absorption edge. Our results are consistent with this paper; ρ_{DB} is reduced upon hydrogenation but it is unchanged by aging and annealing, while $\Delta\theta$ and $\text{tr}(\epsilon)$ are reduced in all three steps as is the optical absorption, indicating that structural relaxation, facilitated by H in the last step, is the most important mechanism in reducing the optical absorption.

Hydrogen in large concentrations is known to reduce the refractive index n of *a*-Si films [40]. The optical characterization of the present films grown at 425 °C with 0.55 at.% H show $n = 3.8$ at 1064 nm and $n = 3.7$

Table I. Bond angle deviation $\Delta\theta$ and local strain $\text{tr}(\epsilon)$ for the *a*-Si films grown at 50, 225 and 425 °C in the as-deposited, annealed (for 3 hours at 425 °C in vacuum) and hydrogenated (for 3 hours at 425 °C in forming gas) states.

	50 °C		225 °C		425 °C	
	$\Delta\theta$ (°)	$\text{tr}(\epsilon)$ (%)	$\Delta\theta$ (°)	$\text{tr}(\epsilon)$ (%)	$\Delta\theta$ (°)	$\text{tr}(\epsilon)$ (%)
as-deposited	10.32 ± 0.09	2.61 ± 0.07	10.05 ± 0.11	1.83 ± 0.05	9.95 ± 0.17	0.77 ± 0.05
annealed	10.09 ± 0.07	1.83 ± 0.05	9.98 ± 0.08	1.61 ± 0.04	9.74 ± 0.11	0.68 ± 0.03
hydrogenated	10.02 ± 0.08	1.70 ± 0.05	9.81 ± 0.09	1.40 ± 0.04	9.70 ± 0.12	0.56 ± 0.03

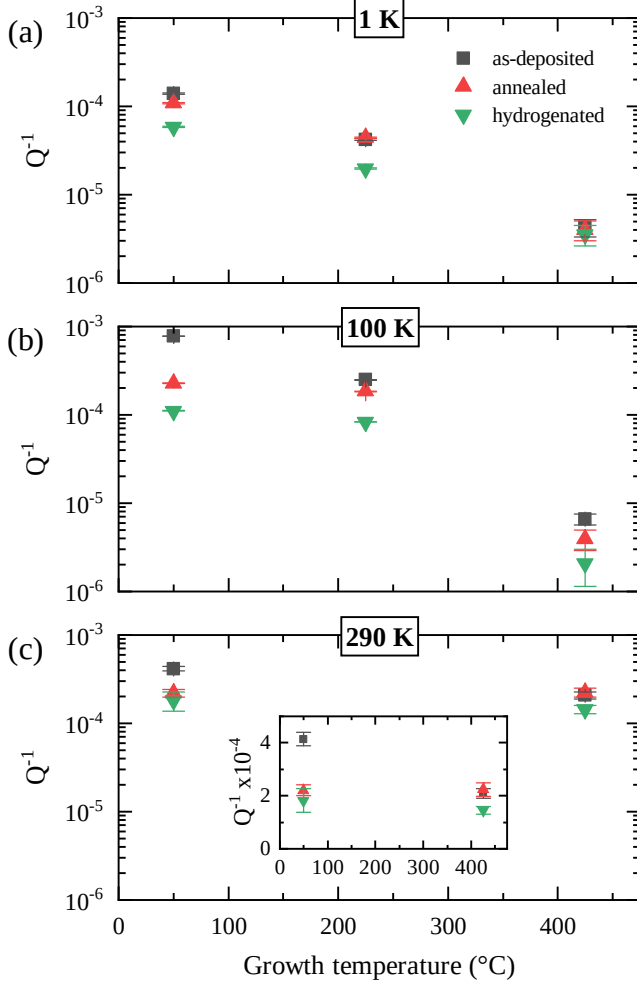


Figure 2. Mechanical loss Q^{-1} of 300 nm *a*-Si films in the as-deposited (black squares), annealed (red upward-pointing triangles), and hydrogenated (green downward-pointing triangles) states as a function of growth temperature. (a) and (b) show Q^{-1} measured by DPOs at 1 K and 100 K, respectively, and (c) shows Q^{-1} measured by GeNS at 290 K. Inset: the same data on a linear scale for increased resolution.

at 2000 nm, and films grown at 50 °C with 1.45 at.% H show $n = 3.7$ at 1064 nm and $n = 3.6$ at 2000 nm (see [25] for more details). These values are comparable to those found in hydrogen-free *a*-Si films at 1064 and 2000 nm, which range from 3.3 to 3.8 [17, 41], and suggests that the at.% H in the present films is too low to

notably reduce their refractive index.

The present plan for Voyager provides a significant sensitivity increase over Advanced LIGO, enabling a substantially increased detection range and event rate [15], but the optical absorption of *a*-Si is not practical. The hydrogenated *a*-Si presented in this work, with an optical absorption of < 3 ppm at 2000 nm for quarter-wavelength-thick films grown at 50 °C, makes Voyager feasible. The sensitivity increase for Voyager in that case will be limited by the mechanical loss of the *a*-SiO₂ layer, not by the *a*-Si layer. As a result, future work will need to focus on developing a low-index material with superior performance compared to *a*-SiO₂, and on optimizing the hydrogenation process of *a*-Si to minimize optical absorption and reduce mechanical loss.

Previous studies on chemical vapor deposited (CVD) *a*-Si:H showed that hydrogen reduces Q^{-1} , but not as a monotonic function of at.% H [42, 43]. These results indicate that preparation conditions contribute to the structural relaxation mechanisms responsible for the Q^{-1} reduction shown for *a*-Si in this letter. Recent research on CVD silicon nitride showed that mechanical loss increases with higher hydrogen levels [20]. Our findings suggest that materials with properties and hydrogen affinity akin to those in *a*-Si may benefit from hydrogenation, reducing mechanical loss when prepared under suitable conditions.

In summary, the optical absorption of *a*-Si shows the largest reduction *and* the lowest values for films grown at 50 °C in the hydrogenated state, which yield an ultra-low optical absorption below our detection threshold of 10 ppm for 500 nm-thick films at 2000 nm, the lowest absorption to date for any *a*-Si. Mechanical loss shows its lowest values of 2×10^{-6} for hydrogenated *a*-Si films grown at 425 °C and measured at 100 K. Hydrogen in *a*-Si reduces dangling bonds and facilitates structural relaxation, where the latter is the most important in reducing optical absorption and mechanical loss.

Future GW detectors, including Voyager and upgrades to LIGO and Virgo, could further increase their sensitivity by combining hydrogenated *a*-Si films with traditional or multimaterial coating designs to reduce thermal noise.

In this letter, we show a trade-off between optimizing optical absorption and mechanical loss with growth temperature. Future studies of hydrogen in *a*-Si should look for optimal preparation conditions. Hydrogenation *during growth* of *a*-Si could potentially yield films with

increased hydrogen concentration and both low optical absorption and mechanical loss. Hydrogenation thus presents a strategy to build silicon-based mirrors for gravitational-wave detectors with lower optical absorption, which is necessary to meet design specifications, and lower mechanical loss, which leads to increased sensitivity, regardless of the wavelength and temperature at which they are designed to operate.

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