Supplemental Material

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

M. Molina-Ruiz ,[1,*] A. Markosyan,[2] R. Bassiri ,[2] M. M. Fejer,[2] M. Abernathy ,[3,†] T. H. Metcalf ,[3] X. Liu ,[3] G. Vajente ,[4] A. Ananyeva,[4] and F. Hellman [1]

[1]*Department of Physics, University of California, Berkeley, California 94720, USA*
[2]*Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA*
[3]*Naval Research Laboratory, Code 7130, Washington, DC 20375, USA*
[4]*LIGO Laboratory, California Institute of Technology, Pasadena, California 91125, USA*

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Preparation and characterization details of the *a*-Si films

Amorphous silicon films 500-nm-thick 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. We have previously done extensive structural analysis on *a*-Si films deposited under the same conditions and shown them to be fully amorphous (by high resolution transmission electron microscopy, Raman spectroscopy and fluctuation electron microscopy) with disorder (specifically bond angle disorder, as measured by Raman peak widths) that decreases and atomic density and sound velocity (shear modulus) that increases with increasing growth temperature [1, 2]. Films were characterized by the techniques described below in their as-deposited, aged, annealed, and hydrogenated states. Films were sequentially aged for 15 months in air, annealed in high vacuum at 425 °C for 3 hours, and hydrogenated in a tube furnace at 425 °C using forming gas (5% H_2 in N_2) for 3 hours.

The hydrogenation of *a*-Si using H_2 benefits from elevated temperature to dissociate the H_2 molecules on the *a*-Si surface and to facilitate H diffusion through the film [3]. To quantify the hydrogen content in the films (resolution of 0.05 at.% H), elastic recoil detection analysis (ERDA) was performed in a NEC model 5SDH Pelletron tandem accelerator, with α particles beam energy of 3040 keV.

The films' thickness was measured using a KLA Tencor ASIQ profilometer, with an error below 1%. The films refractive index n was estimated using a Horiba UVISEL ellipsometer in a spectral range from 300 to 1770 nm at an angle of incidence of 60° and in combination with DeltaPsi2 software to adjust the optical parameters with a Tauc-Lorentz model. Electron paramagnetic resonance (EPR) spectroscopy was used to quantify the dangling bond density ρ_{DB} of the films in the as-deposited and hydrogenated states. EPR was performed at room temperature in a Bruker ELEXSYS E580 EPR spectrometer with an X-band ER 4123D CW-Resonator at 9.36 GHz [4].

These films were confirmed to be amorphous in each state by both Raman spectroscopy and parallel-beam x-ray diffraction. The film's overall disorder, in the as-deposited and hydrogenated states, were evaluated by Raman spectroscopy through determination of the bond angle deviation $\Delta\theta$ and local strain $\mathrm{tr}(\epsilon)$ as discussed elsewhere [2].

The films hydrogenation was done in an atmospheric tube furnace at 425 °C, using a constant forming gas flow with a purity of 99.9999% and a composition of 5% H_2 and 95% N_2. ERDA was performed on *a*-Si films grown at 50 and 425 °C. Figure SM1 shows that the hydrogen depth profile is homogeneous for films grown at 50 °C, while the hydrogen concentration is higher at the surface of films grown at 425 °C.

Figure SM2 shows the ERDA spectra for films grown at 425 °C or hydrogenated for 3 and 60 hours. The results show that the hydrogen concentration and depth profile do not significantly change with annealing time, which suggests that the hydrogenation of homogeneous evaporated *a*-Si films is not a simple diffusion limited process in a homogeneous film.

Figure SM3 shows the refractive index n, measured between 1064 and 2000 nm, of *a*-Si films grown at 50 and 425 °C in the hydrogenated state.

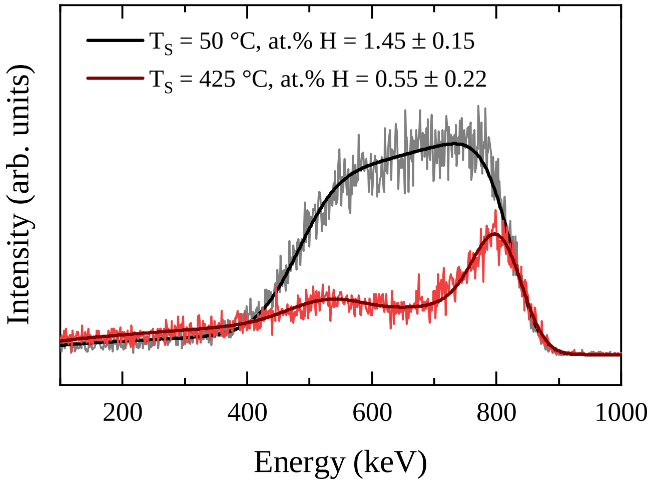

Figure SM1. ERDA spectra of hydrogenated a-Si films grown at 50 and 425 °C (grey and red lines, respectively). Darker smooth lines show the simulation used to determine the films' at.% H.

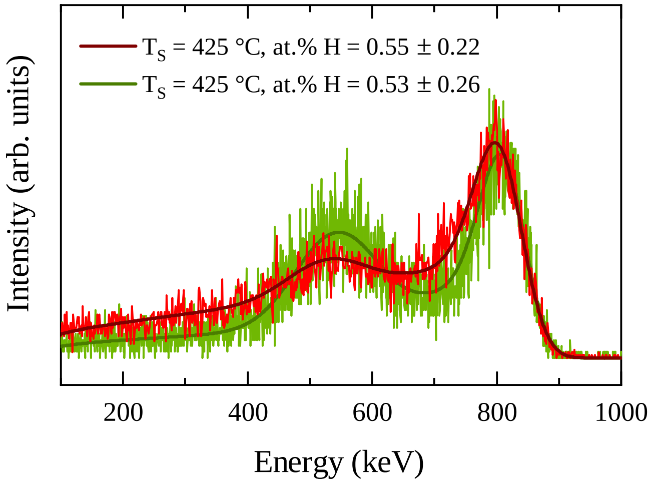

Figure SM2. ERDA spectra of a-Si films grown at 425 °C and hydrogenated for 3 hours (red lines) and for 60 hours (green lines). Darker smooth lines show the simulation used to determine the films' at.% H.

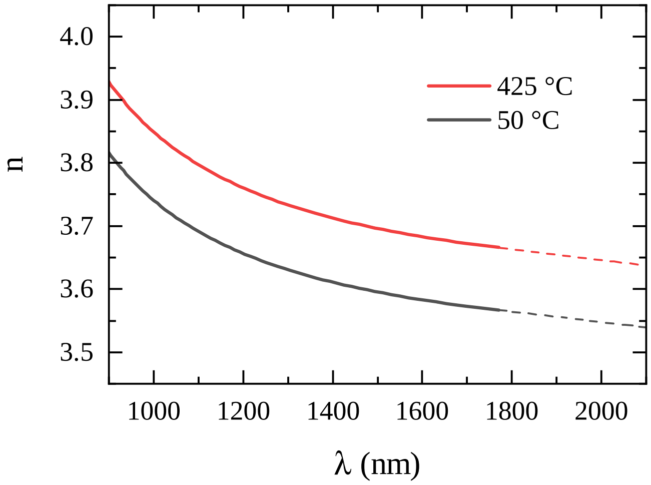

Figure SM3. Refractive index n as a function of wavelength λ for a-Si films in the hydrogenated state and grown at 50 °C (black line) and at 425 °C (red line). Dashed lines show the model extrapolation of the data to obtain n at 2000 nm.

Calculation of the *a*-Si films optical absorption

In the PCI technique, for a film with a refractive index n_f, which is substantially higher than the substrate index n_s, the incident laser beam will set up a standing wave, whose intensity will depend on the reflectivities of both interfaces, and the optical thickness of the film. The intensity will also vary through the thickness of the film, so that appropriate averaging is necessary in order to extract the desired absorption coefficient α. We quantify this effect and correct the measured optical absorption α_m as described below.

The value of the electromagnetic field in the film can be written as

$$E_{e0} = t_1 E_{in} + E_{c0} r_1 r_2 e^{i2\phi} \tag{SM1}$$

where E_{in} and E_{e0} stand for the incident electromagnetic field and the field circulating just inside the top surface of the film. In Eq. SM1, t_1, r_1 and r_2 are the amplitude of the transmission and reflection coefficients for the front (subscript 1) and rear (subscript 2) surfaces, and $\phi = 2\pi nd/\lambda$ is the optical thickness of the film with the physical thickness d. The total power in the film is then

$$P_{tot} = \int_0^d P_{abs}(z)dz = \alpha\frac{1}{2}\int_0^d |E(z)|^2 dz = \alpha d\frac{|E_{in}|^2}{2}(1-r_1^2)F_{corr} \tag{SM2}$$

where the prefactor is the result in the absence of interference effects and the effect of the above interference effect is F_{corr}, given by

$$F_{corr} = \frac{(1+{r_2}^2) + r_2 \sin{(2\phi)}/\phi}{1+{r_1}^2{r_2}^2 - 2r_1 r_2 \cos{(2\phi)}} \tag{SM3}$$

with $r_1 = (n_f - n_0)/(n_f + n_0)$ and $r_2 = (n_s - n_f)/(n_s + n_f)$. In the present case, the refractive indices of the film, substrate and air are $n_f = 3.6$, $n_s = 1.45$ and $n_0 = 1$, respectively. Hence, the measured values of the optical absorption α_m were corrected by F_{corr} yielding the optical absorption values reported in the manuscript given by

$$\alpha = \frac{\alpha_m}{F_{corr}} \tag{SM4}$$

The optical absorption of the RLVIP *a*-Si films discussed in the manuscript is also corrected considering the interference effect described above and divided by a factor of two to account for the different film thicknesses (500 nm vs 1 µm), which enables a quantitative comparison with the present work.

After the measured optical absorption α_m is corrected for the interference effect described above, the absorption coefficient (α/cm) is obtained by dividing α by the film thickness. Since absorption in absolute numbers (in ppm) is most important to the GW community, and mirror coating film thicknesses depend on refractive indices, in the manuscript we quote values for optical absorption.

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[*] manelmolinaruiz@gmail.com

[†] Present address: The Johns Hopkins University Applied Physics Laboratory, Laurel, Maryland 20723, USA

[1] D. Queen, X. Liu, J. Karel, H. Jacks, T. Metcalf, and F. Hellman, J. Non-Cryst. Solids **426**, 19 (2015).

[2] H. C. Jacks, M. Molina-Ruiz, M. H. Weber, J. J. Maldonis, P. M. Voyles, M. R. Abernathy, T. H. Metcalf, X. Liu, and F. Hellman, Phys. Rev. Materials **6**, 045604 (2022).

[3] R. A. Street, in *Hydrogenated amorphous silicon*, edited by R. W. Cahn, E. A. Davis, and I. M. Ward (Cambridge University Press, Cambridge, 1991) 1st ed., Chap. 2, pp. 18–61.

[4] M. Molina-Ruiz, Y. J. Rosen, H. C. Jacks, M. R. Abernathy, T. H. Metcalf, X. Liu, J. L. DuBois, and F. Hellman, Phys. Rev. Materials **5**, 035601 (2021).