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Ultra-low absorption a-Si: beating thermal noise in gravitational astronomy

R. Birney,^{1,2,*} J. Steinlechner,^{3,4,†} Z. Tornasi,³ S. MacFoy,^{1,2} D. Vine,² A. S. Bell,³ D. Gibson,²
J. Hough,³ S. Rowan,³ P. Sortais,⁵ S. Sproules,⁶ S. Tait,³ I. W. Martin,³ and S. Reid^{1,2}

¹*SUPA, Department of Biomedical Engineering, University of Strathclyde, Glasgow, UK*

²*SUPA, Institute for Thin Films, Sensors and Imaging,
University of the West of Scotland, Paisley, UK*

³*SUPA, Institute for Gravitational Research, University of Glasgow, UK*

⁴*Institut für Laserphysik and Zentrum für Optische Quantentechnologien,
Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany*

⁵*Polygon Physics, 30 Chemin de Rochasson, 38240, Meylan, France*

⁶*WestCHEM, School of Chemistry, University of Glasgow, UK*

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Amorphous silicon has ideal properties for many applications in fundamental research and industry. However, the optical absorption is often unacceptably high, particularly for gravitational wave detection. We report a novel ion beam deposition method for fabricating amorphous silicon with unprecedentedly low unpaired electron spin density and optical absorption; the spin-limit on absorption being surpassed for the first time. At low unpaired electron density, the absorption is no longer correlated with electron spins, but with the electronic mobility gap. Compared to standard ion beam deposition, the absorption at 1550 nm is lower by a factor of ≈ 100 . This breakthrough shows that amorphous silicon could be exploited as an extreme performance optical coating in near-infra-red applications and it represents an important proof-of-concept for future gravitational wave detectors.

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Introduction – Highly-reflective optical coatings have a wide range of applications in research and technology. Ultrastable optical cavities are essential components in atomic clocks, which are revolutionizing time and frequency standards and measurement [1–3]. Ultrastable cavities also form the heart of a gravitational-wave detector. The measurement of gravitational waves is an exciting tool for astrophysics, making dark objects such as black holes visible [4–7]. In all of these applications, performance is currently limited by Brownian thermal noise, which is proportional to the mechanical loss and thickness of the mirror coatings [8–11].

Amorphous silicon (a-Si) is a highly interesting coating material due to low mechanical loss at room temperature, which decreases towards low temperatures [12, 13], and a very high refractive index of approximately $n = 3.5$ in the NIR. Highly-reflective dielectric mirror coatings comprise alternating layers of materials with low and high n . Typically, the layers are a quarter of the design wavelength in optical thickness (QWOT); optical thickness being equal to nd , where d is physical thickness of the layer; two of the most commonly-used wavelengths being 1064 nm and 1550 nm. Compared to materials of lower n , the high index of a-Si allows fewer layers to be deposited in order to achieve the same reflectivity, due to a higher refractive index contrast Δn between the two materials. Additionally, the quarterwave thickness is directly reduced.

To avoid heating and thermal deformation of the mirrors in gravitational wave detectors, or to realise ultra-high finesse cavities, low optical absorption at the ppm (10^{-6}) level is required. However, the optical absorp-

tion of a-Si may be significantly higher [14]. Recent research has resulted in an absorption reduction of more than a factor of 50 when using a-Si at a wavelength of $2\ \mu\text{m}$, and at low temperatures [15, 16]. However, shorter wavelengths are preferable, since an increase in wavelength increases the coating thickness by the ratio of the wavelengths, and therefore coating thermal noise by the square root of the ratios. In addition, the telecommunication wavelength of 1550 nm is attractive, due to the ready availability of high power lasers and optical components.

Incorporating hydrogen into a-Si has been reported to significantly reduce optical absorption [17]. However, hydrogenation may be undesirable due to reduction of the refractive index and may result in the formation of infrared absorbing hydroxyl (OH) groups when combined with frequently used low- n oxide materials (e.g. SiO_2).

In this Letter, we describe a novel ion-beam deposition (IBD) process for fabricating hydrogen-free low-absorbing a-Si coatings. We show that it is possible to reduce the number of unpaired electrons to a level at which they no longer significantly contribute to absorption. In this regime, absorption remains correlated with the electronic mobility gap. We investigate the optimum heat-treatment temperature and the effect of elevated temperature deposition on the material. The optical absorption reaches a minimum upon heat treatment at 400°C , while mechanical dissipation at room temperature is minimized by deposition at 200°C , followed by post-deposition heat treatment at 400°C .

The lowest absorption achieved corresponds to an ex-

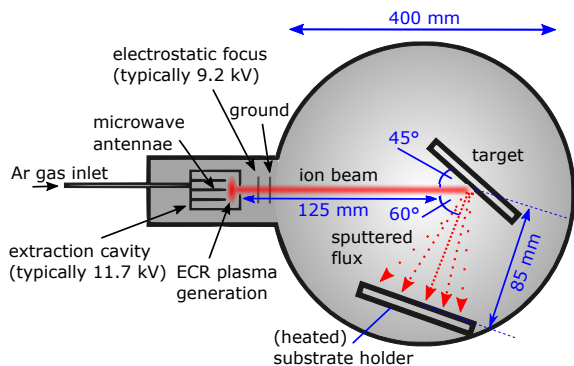


FIG. 1. Schematic of the deposition setup for producing ultra-low absorbing a-Si.

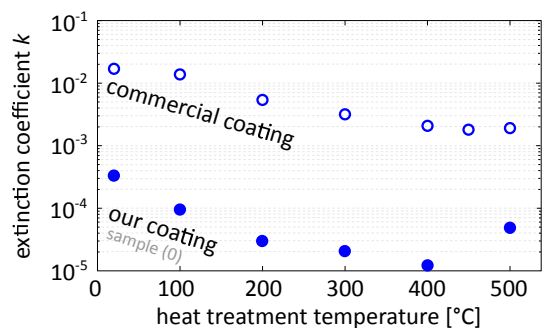


FIG. 2. Extinction coefficient k at 1550 nm as a function of heat treatment temperature for our coating and, for comparison, of a commercial coating (data from [15]).

86 extinction coefficient of $k = (1.2 \pm 0.2) \times 10^{-5}$ at 1550 nm¹²⁵
 87 and of $k = (1.7 \pm 0.1) \times 10^{-4}$ at 1064 nm. This is¹²⁶
 88 approximately 25 \times lower at 1064 nm, and more than¹²⁷
 89 100 \times lower at 1550 nm [15], than previously reported for¹²⁸
 90 IBD-deposited thin films. ¹²⁹

91 *Coating Deposition* – IBD is commonly used to pro-
 92 duce the highest-quality optical coatings with low opti-¹³¹
 93 cal absorption and scatter. The a-Si coatings investi-¹³²
 94 gated here were produced by a custom-built IBD system¹³³
 95 (see Fig. 1), incorporating a novel electron cyclotron res-¹³⁴
 96 onance (ECR) ion source [18]. ¹³⁵

97 The ion beam is formed by injection of argon gas¹³⁶
 98 into a resonant microwave cavity where it is ionized via¹³⁷
 99 ECR [19]. The cavity was tuned to 2.45 GHz and the¹³⁸
 100 microwave power was held constant at 11.6 W. In con-¹³⁹
 101 ventional IBD, the cavity walls are held at high voltage¹⁴⁰
 102 and the ions are extracted through a grid. The higher¹⁴¹
 103 frequency of ECR sources [20, 21] enables generation of¹⁴²
 104 a more highly-confined plasma, which can be extracted¹⁴³
 105 through a single aperture. This reduces the possibility¹⁴⁴
 106 of contamination from the grid material and permits ex-¹⁴⁵
 107 traction potentials an order of magnitude larger (11.7 kV¹⁴⁶
 108 in this study). ¹⁴⁷

109 The deposition rate used here of $\sim 0.05 \text{ \AA/s}$ is \approx ¹⁴⁸
 110 20 times lower than for conventional IBD. Deposition¹⁴⁹
 111 rate is known to affect atomic structure during thin film¹⁵⁰
 112 growth [22, 23], and therefore may play an important role¹⁵¹
 113 in reducing the density of under-coordinated Si atoms. ¹⁵²

114 a-Si coatings were deposited using an N-type¹⁵³
 115 (phosphorus-doped) crystalline silicon (semiconductor¹⁵⁴
 116 grade) target with resistivity = 1 – 10 Ωcm . Base pres-¹⁵⁵
 117 sure in the chamber prior to deposition was a maxi-¹⁵⁶
 118 mum of 1×10^{-6} mbar (averaging 5×10^{-7} mbar), and¹⁵⁷
 119 8×10^{-5} mbar during deposition. Coatings were de-¹⁵⁸
 120 posited in a newly-built vacuum chamber; no other coat-¹⁵⁹
 121 ing materials had previously been produced in this sys-¹⁶⁰
 122 tem and the deposition environment was therefore largely¹⁶¹
 123 free of potential contaminants. Elemental analysis was¹⁶²
 124 conducted via energy-dispersive X-ray spectroscopy, us-¹⁶³

ing room temperature-deposited a-Si films on GaAs sub-
 strates. The oxygen content was quantified to be $\leq 5\%$,
 consistent with that expected from the slow deposition
 rate and base pressure in the coating chamber. SiH and
 SiH₂ content was estimated to be $< 1\%$ with Raman
 spectroscopy [27].

Optical Absorption Measurements – Substrates made
 of Corning 7979 [24] and JGS-1 [25] fused silica,
 which show negligible optical absorption at 1064 nm and
 1550 nm, were coated for absorption measurements. Dur-
 ing the coating process, the substrates were mounted on a
 stage with heating capability. Coatings were deposited at
 room temperature (with an initial substrate temperature
 of 20 °C, increasing to 35 °C after 1 h deposition), and at
 elevated substrate temperatures of 200 °C and 400 °C.

Optical absorption of the a-Si films was measured using
 photothermal common-path interferometry (PCI) [26].
 Accounting for interference effects, the extinction coef-
 ficient k was calculated [28].

Figure 2 shows k at 1550 nm of a room-temperature de-
 posited a-Si sample as a function of post-deposition heat-
 treatment temperature. The sample was heat treated for
 1 hr in air for each heat-treatment step. k shows a mini-
 mum of $(1.22 \pm 0.21) \times 10^{-5}$ after heat treatment at 400 °C
 . This corresponds to an absorption of a highly-reflective
 a-Si/SiO₂ stack of (7.6 ± 1.4) ppm, assuming negligible
 absorption in the SiO₂ layers [15]. A commercial a-Si
 coating produced via IBD by *Advanced Thin Films* is
 shown for comparison (data from [15]).

Figure 3 shows k at 1550 nm as a function of deposition
 temperature. Each sample was measured after deposition
 and then heat-treated at 400 °C for 3 hours (except for
 the points taken from Fig. 2). For room temperature de-
 position, k shows a wide spread for nominally identical
 deposition parameters. However, on average, a general
 decreasing trend of k with deposition temperature is ob-
 servable for the as-deposited samples, and all individual
 samples show a decrease in k following heat treatment.
 We note that post-deposition heat-treatment can result

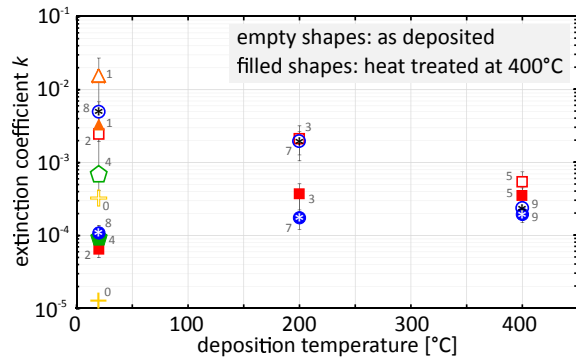


FIG. 3. Extinction coefficient k at 1550 nm as a function of deposition temperature. At each temperature, different coatings are indicated by different shapes. (Crosses represent our coating from Fig. 2; stars indicate coatings deposited on Corning 7979 substrates. All other coatings were deposited on JGS-1 substrates.

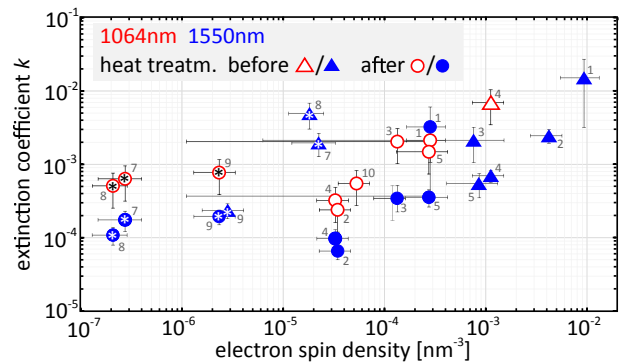


FIG. 4. Extinction coefficient k at 1064 nm (red) and 1550 nm (blue) of a-Si coatings as a function of electron spin density. Stars (*) indicate coatings on Corning 7979 substrates, all other coatings were on JGS-1.

164 in lower k values than elevated temperature deposition¹⁹⁶
 165 at the same temperature alone. The improvement with¹⁹⁷
 166 post-deposition heat-treatment at deposition tempera-¹⁹⁸
 167 ture is small. We assume that the spread in absorption¹⁹⁹
 168 for films deposited under nominally identical conditions²⁰⁰
 169 arises from an unknown variation in deposition parame-²⁰¹
 170 ters, most likely chamber cleanliness. Since the coatings²⁰²
 171 with the lowest absorption were among the first produced²⁰³
 172 in the IBD system following commissioning and testing,²⁰⁴
 173 it seems likely that absorption variations may be related²⁰⁵
 174 to accumulating contamination of the coating chamber.²⁰⁶

175 *Optical absorption mechanisms* – Unpaired electrons²⁰⁷
 176 are known to contribute to the absorption in a-Si [29].²⁰⁸
 177 The density of unpaired electrons (‘spin density’) of several²⁰⁹
 178 samples was measured via electron paramagnetic resonance²¹⁰
 179 (EPR) [30]. Figure 4 shows k versus number of²¹¹
 180 electron spins per nm^3 for a variety of samples, some of²¹²
 181 which were deposited at room temperature, some at ele-²¹³
 182 vated temperature and some were heat treated at 400 °C²¹⁴
 183 after deposition. The absorption was measured for the²¹⁵
 184 same samples at both 1064 nm and 1550 nm¹, and we²¹⁶
 185 note the evidence of substrate effects in these measure-²¹⁷
 186 ments which warrants further investigation.²¹⁸

187 Both heat treatment and high temperature deposition²¹⁹
 188 can be observed to reduce the spin density, in addition²²⁰
 189 to the previously noted reduction in absorption. Sam-²²¹
 190 ples 4 and 9, which were deposited and heat-treated at²²²
 191 400 °C, show little or no significant change in spin density²²³
 192 following heat-treatment – consistent with the minimal²²⁴
 193 reduction in absorption in these samples following heat²²⁵
 194 treatment at deposition temperature. When considering²²⁶
 227

all samples, a decrease in k with decreasing spin density is observed for spin densities above $\approx 4 \times 10^{-5}/\text{nm}^3$, with broadly linear dependence, in good agreement with other studies [17]. However, we observe that when the spin density is reduced below $\approx 4 \times 10^{-5}/\text{nm}^3$, no further decrease in absorption is observed. This indicates that another absorption mechanism dominates in this regime. It is interesting to note that the spin density typically observed in non-hydrogenated a-Si [31, 32] is in the order of $5 \times 10^{-3} \text{ nm}^{-3}$, significantly higher than observed in the majority of our ECR-IBD films.

The relationship between absorption and electronic structure in the low spin density regime in Fig. 4 was investigated through analysis of the a-Si coatings’ transmittance spectra between 200–2000 nm.

Spectra were analyzed using the software package SCOUT [33], with the dielectric function of a-Si modeled as the sum of a constant dielectric background [34], an ‘OJL’ term [35] to model inter-band transitions, and an extended Drude term [36] representing electron transport properties. The dielectric function of the substrate was calculated separately, allowing the total transmittance of a-Si on fused silica to be modeled and fitted to the measured spectrum.

The fitting parameter of interest to this study is the OJL mobility gap, E_g , which is related to the position of the transmittance-spectrum absorption-edge. The localized-state decay-constants were taken to be identical for the valence and conduction bands ($\gamma_{\text{val}} = \gamma_{\text{cond}}$). The lowest optical absorption is observed in the ‘plateau’ region not dominated by electron spins in Fig. 4. A correlation is suggested between extinction coefficient and mobility gap (Fig. 5), in agreement with the hypothesis that the mechanism for absorption is inter-band transitions rather than absorption by defects, impurities or dangling bonds. No correlation was observed with γ , indicative of the degree of disorder (there are various types and degrees of disorder that are known to affect the mo-

¹ Several samples were not measured at 1064 nm before heat treat-²³⁰
 ment, as they had already been heat treated for the 1550 nm²³¹
 measurements.²³²

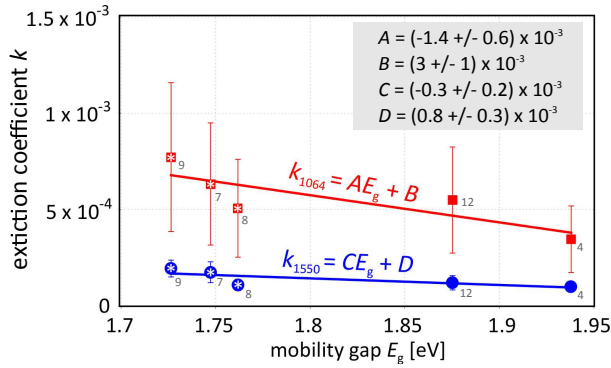


FIG. 5. Extinction coefficient k as a function of the calculated mobility gap energy from the OJL model for absorption results in the plateau region of Figure 4, with linear fit. Stars indicate coatings on Corning 7979 substrates, all other coatings were on JGS-1.

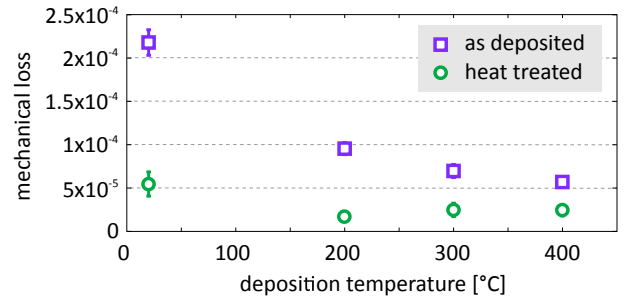


FIG. 6. Coating mechanical loss as a function of deposition temperature: Each point represents the average loss of several resonant modes (purple squares: coating as-deposited, green circles: heat treatment for 1 hr at 400°C).

TABLE I. Thermal noise possible when using ECR-IBD a-Si in a multi-material coating compared to Advanced LIGO. The numbers are for ETMs with a beam diameter of 6.2 cm and in brackets for the ITMs with a beam diameter of 5.5 cm as used in Advanced LIGO. Mechanical loss values used for Ti:Ta₂O₅ and SiO₂ are taken from [40].

thermal noise*	absorption	no. bilayers ETM (ITM)	
[%]	[ppm]	Ti:Ta ₂ O ₅ /SiO ₂	a-Si/SiO ₂
baseline Advanced LIGO (a)			
100	≈ 0.3 (0.2) [43]	18.5 (9.5)	0 (0)
a-Si/SiO ₂ 1550 nm (b)			
29.9	7.6	-	7.5 (4.5)
multi-material 1550 nm (c)			
49.5	2.1 (2.0)	2 (2)	6.5 (2.5)

*for whole detector

bility gap edges and in a-Si [37]). The value of γ obtained from all fits was very similar, with an average value of 0.12 ± 0.02 .

It is known that E_g for an amorphous semiconductor decreases as the average atomic spacing increases [38]. Thus, a further decrease in this remarkably low absorption may be possible through decreasing the average atomic spacing via optimization of deposition parameters; specifically, increased extraction potential, *i.e.* higher ion energy (see *Coating Deposition* Section for parameters used), or the incorporation of addition processes known to improve densification *e.g.* ion assist.

Thermal noise performance – To estimate the thermal noise performance of these coatings, fused silica cantilevers were coated at the same temperatures as the disc samples, to facilitate studies of the mechanical loss. Coating mechanical loss may be calculated from the difference between the free amplitude decay of the cantilevers' resonant modes before and after coating [39].

Figure 6 shows the coating mechanical loss as a function of deposition temperature. The purple squares show the average loss of several bending modes of the as-deposited coating and the green circles show the average loss of the coating after heat treatment at 400°C. The lowest coating loss of $\phi = (1.7 \pm 0.1) \times 10^{-5}$ was found for deposition at 200 °C followed by post-deposition heat treatment at 400 °C. No frequency dependence was observed, with the losses approximately a factor of 5 lower than that previously reported for identically treated a-Si coatings deposited by conventional IBD [12].

Table I compares thermal noise for different coatings used in the Advanced LIGO detectors. The total thermal noise has contributions from two cavity input mirrors (ITMs) and two cavity end mirrors (ETMs). Thermal noise of the current Advanced LIGO coatings, consisting of Ta₂O₅ doped with TiO₂ (Ti:Ta₂O₅) and SiO₂ at a 290

wavelength of 1064 nm (coating (a)), is defined as 100%. Using SiO₂ together with the lowest absorption and mechanical loss found for our a-Si at 1550 nm (coating (b)) reduces thermal noise to 29.9% that of coating (a) for similar mirror transmissions.

While being remarkably low for a-Si, the absorption of 7.6 ppm is still above the tolerable level for use in gravitational wave detectors. In the silica Advanced LIGO mirrors, tolerable levels of thermal distortion may suggest a maximum coating absorption of 2.5 ppm [41, 42]. A method of further reducing the absorption of coating (b) is a 'multi-material' design, in which low-absorbing Ti:Ta₂O₅/SiO₂ layers on top of the coating reduce the laser power before it arrives at the a-Si layers [44, 45]. Depending on the number of Ti:Ta₂O₅/SiO₂ layers, absorption in the a-Si may be tuned. However, this tuning requires a trade-off between absorption reduction and thermal-noise increase due to the higher mechanical loss of Ti:Ta₂O₅/SiO₂. Using two bilayers of Ti:Ta₂O₅/SiO₂ reduces the absorption to < 2.5 ppm, with a slight increase in thermal noise to 49.5% of coating (a). This meets the Advanced LIGO Plus requirement of a factor of two reduction in thermal noise [46].

Conclusion – We have developed a process for depositing hydrogen-free a-Si films with unprecedentedly low electron spin density. The absorption is correlated with the electron-spin density for densities above $\approx 1 \times 10^{-5}/\text{nm}^3$, below which it is correlated with the electronic mobility gap. Films with optical absorption a factor of ≈ 100 lower at 1550 nm ($\approx 25 \times$ lower at 1064 nm) than for conventional IBD a-Si have been produced. The mechanical loss after optimal heat-treatment is $\approx 5 \times$ lower than for a-Si deposited by conventional IBD.

The very low optical absorption and mechanical loss enable the use of a-Si for significant thermal noise reduction in precision measurements. A multi-material design can reduce coating thermal noise to 49.5% of the Advanced LIGO level, for a change in wavelength to 1550 nm, while keeping the absorption < 2.5 ppm. This provides, for the first time, a route to significant sensitivity improvement at room temperature, exceeding the requirements for the planned Advanced LIGO Plus detector [46], designed to increase detection rates by a factor of ≈ 5 .

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* Ross.Birney@strath.ac.uk

† Jessica.Steinlechner@ligo.org

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