



1 Supporting Information

Preferential Orientation of Photochromic Gadolinium
 Oxyhydride Films

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## 18 Optical Characterization

Figure S1 shows (a) optical transmittance T and (b) absorptance A as a function of wavelength  $\lambda$ and photon energy E, respectively, for photochromic GdHO samples (clear state) deposited at pressures P ranging from 1.5 to 5.8 Pa. As P increases, T increases in the visible regime between 400 and 700 nm (a) and the absorption edge, located at shorter wavelengths, shifts towards higher energies (b).





Figure S2 shows the difference in (a) transmittance  $\Delta T$ , (b) absorptance  $\Delta A$  and (c) reflectance AR for films deposited at different P before and after illumination. The absorption edge shifts towards shorter wavelengths with increasing P indicating a widening of the bandgap (bandgap energies are tabulated in Table S1).  $E_g^{dir}$  increases from 2.8 to 3.7 eV as P increases from 1.5 to 5.8 Pa.

Table S1. Bandgap values of samples deposited between 1.5 Pa and 5.8 pa.; where Egdir refers to the
 energy of the direct bandgap.

Sample	Deposition	Film Thickness	$E_{g^{dir}}$
	pressure (Pa)	(nm)	(eV)
GdH₃			2.4[1]
GdHO	1.5	616	2.8
	2.0	575	3.0
	2.8	550	3.1
	3.8	525	3.3
	5.8	540	3.7
Gd <sub>2</sub> O <sub>3</sub>			5.4[2]

Figure S2 (d) shows the absorptance averaged between 550 nm and 1000 nm in the clear A<sub>clear</sub> and photodarkenened A<sub>dark</sub> states, as well as the photochromic response  $|\Delta A|$  as a function of deposition pressure. The photochromic response decreases as P increases (e.g., P = 1.5 Pa,  $|\Delta A| = 47$ % and P = 5.8 Pa,  $|\Delta A| \approx 0$  %). These results, which are consistent with previous reports [3], are attributed to the increase of oxygen content in the films prepared at larger P, resulting in higher porosity [4], favoring thus the exchange of hydrogen and oxygen once the samples are removed from the sputtering chamber and exposed to ambient conditions [5].





Figure S2. Change in (a) transmittance, (b) reflectance and (c) absorptance of samples plotted versus
 wavelength, deposited between 1.5 Pa and 5.8 Pa before and after 60 minutes of illumination. (d)
 Absorptance and photochromic response, averaged between 550-1000 nm, of samples plotted versus
 pressure.

## 46 Composition analysis

Figure S3 (a and b) shows normalized and interpolated maps of the O and H content as a function of deposition pressure and depth. C impurities of up to 4 at. % are not indicated. All the majority elements (i.e., Gd, O and H) are uniformly distributed throughout the films, except at the surface, where an oxygen-rich layer (within 5-10 nm) is found. The increase in deposition pressure

<sup>33</sup> 

51 leads to stronger oxidation and decrease of hydrogen content. This effect can be attributed to 52 variations of the porosity of the films [4]. The replacement of H atoms by O atoms during the 53 oxidation process is suggested by the anti-correlation in the O and H content (Figure S3). Typical 54 uncertainties - statistical and systematic - involved in this measurement, especially towards lighter 55 elements as H, are discussed in detail using similar system elsewhere [6].

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57 **Figure S3.** Depth profiles of (a) hydrogen and (b) oxygen plotted versus deposition pressure 58 deduced from Tof-E ERDA coincidence spectra (not whown).

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75