nanomaterials

Supplementary Materials

Hysteresis-Free Near-Infrared Optical Hydrogen Sensor Based on Ti/Pd Bilayer Thin Films

Ashwin Thapa Magar [1], *, Tu Anh Ngo [1], Hoang Mai Luong [2], Thi Thu Trinh Phan [3], M. Tuan Trinh [3], Yiping Zhao [1], and Tho D. Nguyen [1], *

[1] Department of Physics and Astronomy, University of Georgia, Athens, GA 30602, USA
[2] Department of Electrical Engineering, Chulalongkorn University, Bangkok 10330, Thailand
[3] Department of Chemistry and Biochemistry, Utah State University, Logan, UT 84322, USA
* Correspondence: ashwin.thapa@uga.edu, ngtho@uga.edu

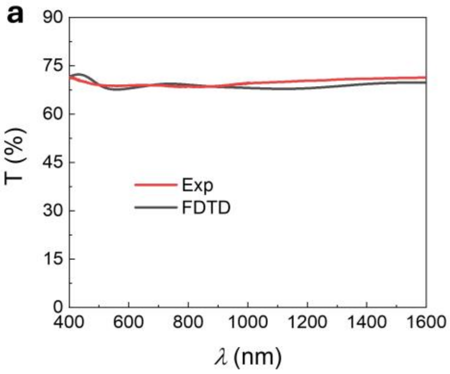
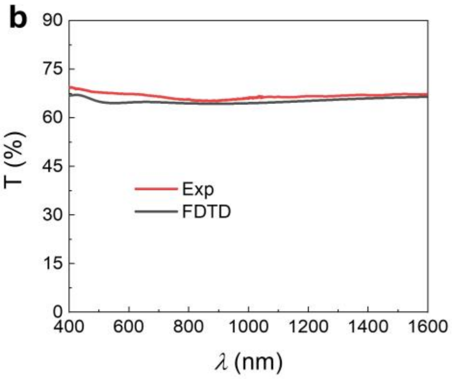


Figure S1. Comparison between experimental (Exp) and finite-difference time-domain (FDTD) simulated optical transmission spectra for (a) 2.5 nm Pd/TAF, and (b) 5 nm Ti/2.5 nm Pd/TAF thin film configurations.



Academic Editor:

Received: date
Revised: date
Accepted: date
Published: date

Citation: To be added by editorial staff during production.

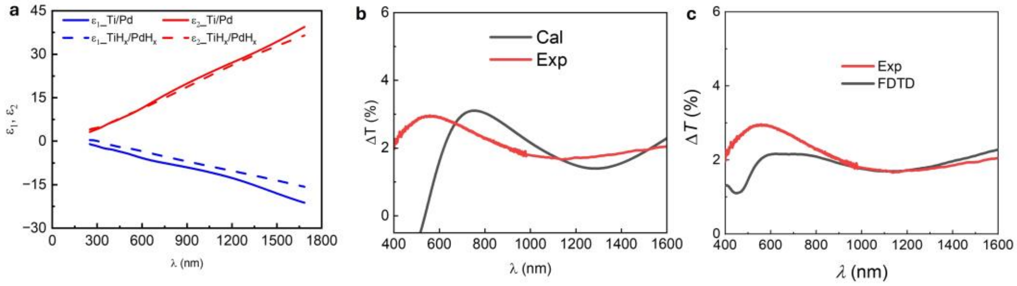


Figure S2. (a) Dielectric functions of the metal and its hydride for the 5 nm Ti/2.5 nm Pd bilayer structure, calculated using S1 equation. (b) Hydrogen-induced optical transmission change (ΔT) obtained from experimental measurements (Exp) and theoretical calculations (Cal) based on the equation 1 described in the main text. (c) Comparison between simulated (FDTD) and experimental ΔT spectra for the same bilayer sample.

Dielectric functions of the metals Pd and Ti along with their respective hydrides PdH_x and TiH_x were extracted from literature data [1] . To estimate the optical behavior of the bilayer (5 nm Ti/2.5 nm Pd) structure, the effective dielectric function was calculated using a weighted average approach (Eq. s1), appropriate for thin films whose total thickness is much smaller than the wavelength of interest [2]. The expression for the effective dielectric function is given by:

$$\varepsilon_{\mathrm{eff}} = \frac{d_{\mathrm{Pd}} \times \varepsilon_{\mathrm{Pd}} + d_{\mathrm{Ti}} \times \varepsilon_{\mathrm{Ti}}}{d_{\mathrm{Ti}} + d_{\mathrm{Pd}}} \qquad \text{(S1)}$$

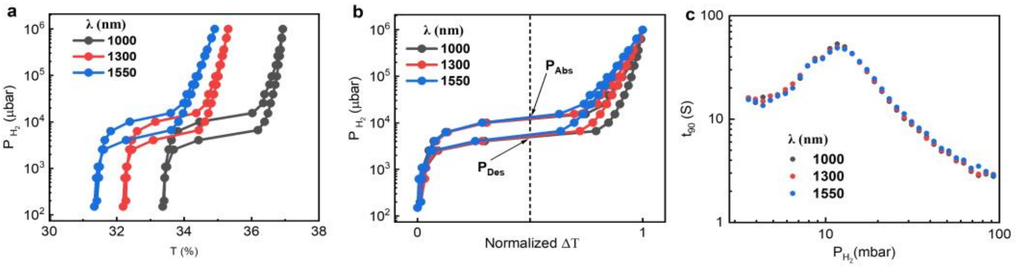


Figure S3. (a) Optical hydrogen sorption isotherms of the 5 nm thin film measured at various near-infrared (NIR) wavelengths. (b) Normalized isotherms corresponding to the wavelengths shown in panel (a). (c) Hydrogen response times at the corresponding NIR wavelengths.

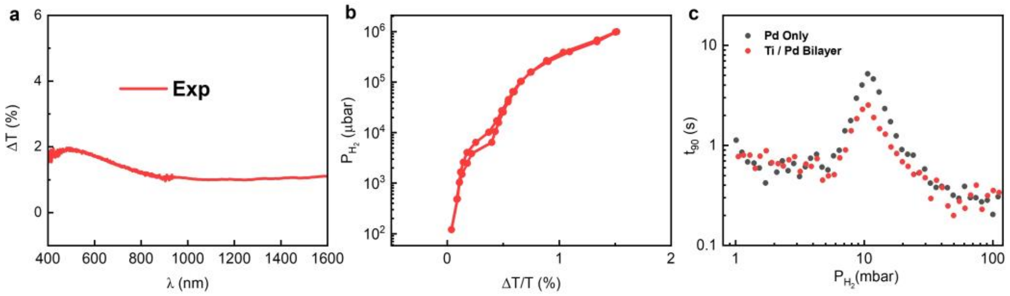


Figure S4. Sensing performance of the 5 nm Ti/ 2.25 nm Pd/ 30 nm TAF sensor (a) Hydrogen-induced optical transmission changes ($\Delta T(\lambda) = T_{1000\ \mathrm{mbar}} - T_{0\ \mathrm{mbar}}$) across the spectral range of 400 – 1600 nm. (b) Optical hydrogen sorption isotherm at $\lambda = 1550$ nm. (c) Comparison of response times between the Ti/Pd/TAF sensor and Pd/TAF reference film under hydrogen pressure ranging from 1 to 100 mbar.

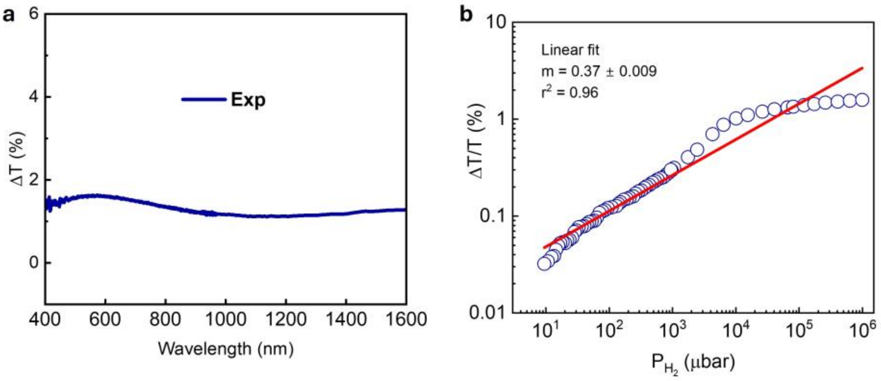


Figure S5. Optimized 5 nm Ti/ 1.9 nm Pd/ 30 nm TAF sensor (a) Hydrogen-induced optical transmission changes ($\Delta T(\lambda) = T_{1000\ \mathrm{mbar}} - T_{0\ \mathrm{mbar}}$) of the across the 400 – 1600 nm spectral range (b) Limit of detection (LOD) of the optimize sensor, where the sensor response follows sievert's power law, expressed as, $\frac{\Delta T}{T} \propto (P_{H_2})^m$ [3].

References

1. Palm, K.J., et al., *Dynamic Optical Properties of Metal Hydrides.* ACS Photonics, 2018. **5**(11): p. 4677-4686.
2. Hu, F.G., J. Song, and T. Kamgaing. *Modeling of multilayered media using effective medium theory*. in *19th Topical Meeting on Electrical Performance of Electronic Packaging and Systems*. 2010.
3. Caravella, A., et al., *Sieverts Law Empirical Exponent for Pd-Based Membranes: Critical Analysis in Pure H2 Permeation.* The Journal of Physical Chemistry B, 2010. **114**(18): p. 6033-6047.