**Electronic Supplementary Information** 

## Synergistic enhancement of hydrogen interactions in palladium-gold-silicon metallic glass on multilayered graphene<sup>†</sup>

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**Fig. S1.** Tribological analysis of the (a) PdSiAu+Si/SiO<sub>2</sub> and (b) MLGR+PdSiAu+Si/SiO<sub>2</sub> using atomic force microscopy. The room mean square (RMS) roughness, projected and active areas are provided below the corresponding images.



**Fig S2.** Raman spectroscopy of MLGR+PdSiAu+Si/SiO<sub>2</sub>, PdSiAu+Si/SiO<sub>2</sub> and Si/SiO<sub>2</sub> showing the full spectra.



**Fig. S3.** Plot of  $1/It^{1/2}$  vs. *t* for MLGR+PdSiAu+Si/SiO<sub>2</sub>. The open blue squares and the red line depict measured data and the best linear fit. The slope and y-intercept of the linear fit correspond,

respectively, to  $\frac{R_{\Sigma}}{\Delta E} = \frac{l\sqrt{\pi}}{\Delta Q\sqrt{D}}$  in equation 1. Linear fitting is applied to the region where the data are linearized until the saturation point.



Fig. S4. Plot of  $1/It^{1/2}$  vs. t for PdSiAu+Si/SiO<sub>2</sub>. The open blue squares and the red line depict measured data and the best linear fit. The slope and y-intercept of the linear fit correspond,

$$\frac{R_{\Sigma}}{1}$$
  $\frac{l\sqrt{\pi}}{1000}$ 

respectively, to  $\overline{\Delta E}$  and  $\Delta Q \sqrt{D}$  in equation 1. Linear fitting is applied to the region where the data are linearized until the saturation point.



**Fig. S5.** HRTEM imaging of the as-sputter deposited MLGR+PdSiAu layers and FFT pattern of the region indicated by the cyan square.



**Fig. S6.** Close-up HRTEM image of the MGTF -  $Si/SiO_2$  interface showing PdH<sub>x</sub> nanocrystals.



**Fig. S7.** HRTEM imaging of the entire layer of the as-sputter deposited MLGR+PdSiAu layers on Si/SiO<sub>2</sub> substrate and FFT pattern of the region indicated by the light green square.



Fig. S8. HRTEM imaging of the PdSiAu MGTF and Si/SiO $_2$  interface



**Fig. S9.** HAADF imaging after hydrogenation of MLGR+PdSiAu+Si/SiO<sub>2</sub>; scattered black regions correspond to nanovoids formed during hydrogen-metal interactions.