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Palladium and gold nanotube arrays with tuned surfaces for low-temperature hydrogen sensing

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Synthesis of AAO templates

The synthetic procedures of AAO templates are as follows. [1, 2] Firstly, high-purity aluminum foils (99.999%) are annealed at 500 °C for 5 hours to eliminate internal stress and improve plasticity. Secondly, the first anodization is conducted in 0.3 M oxalic acid solution at 10 °C under a constant voltage of 40 V for 4 hours. Thirdly, after removing the formed oxide layer in a mixed phosphoric-chromic acid solution, a second anodization is performed under the same conditions for 16 hours to obtain well-ordered nanochannels. Fourthly, the residual Al layer on the opposite side of the AAO is removed using a $SnCl_4$ solution. Fifthly, the obtained template is immersed in 5 wt% phosphoric acid to obtain through-hole AAO. Finally, the AAO template was rinsed thoroughly with deionized water, dried completely, and sputtered with a thin Au layer to obtain the final AAO template.

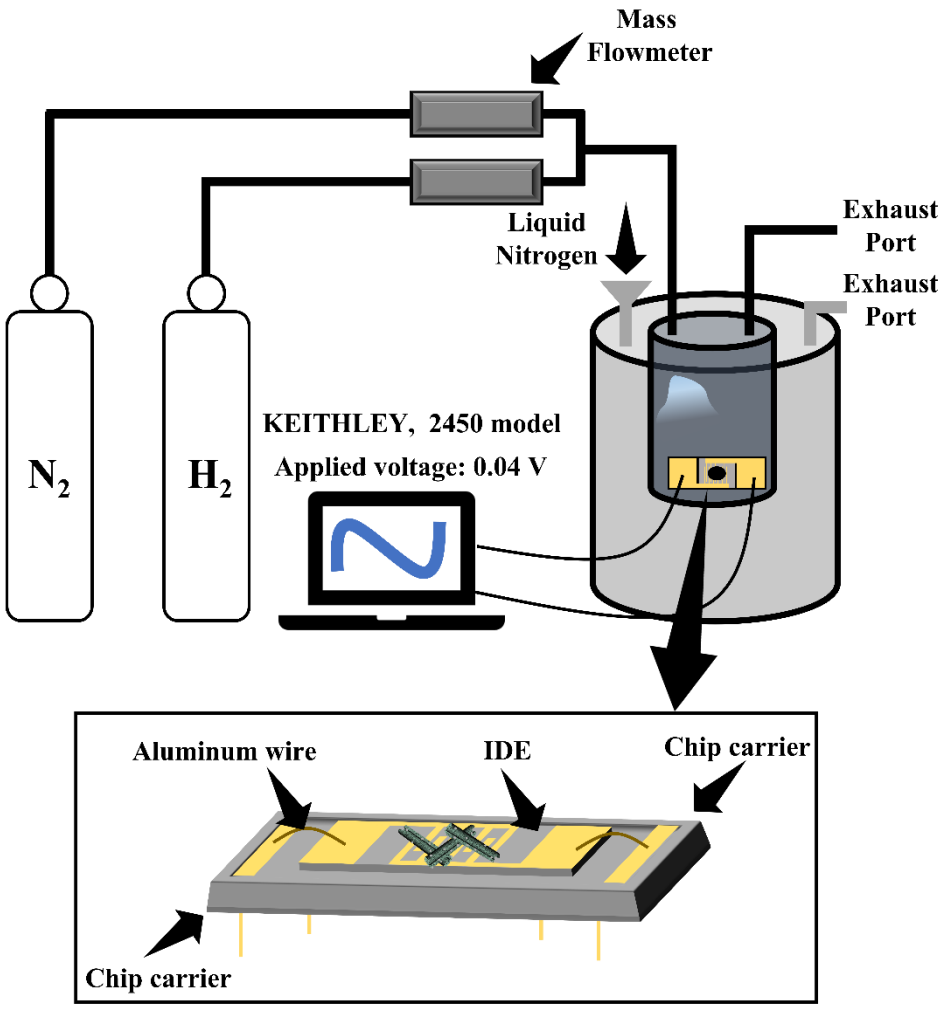

Fig. S1 The schematic diagram of the evaluation setup for H_2 sensing and the sensor prototype built with RB-PdAu NTs.

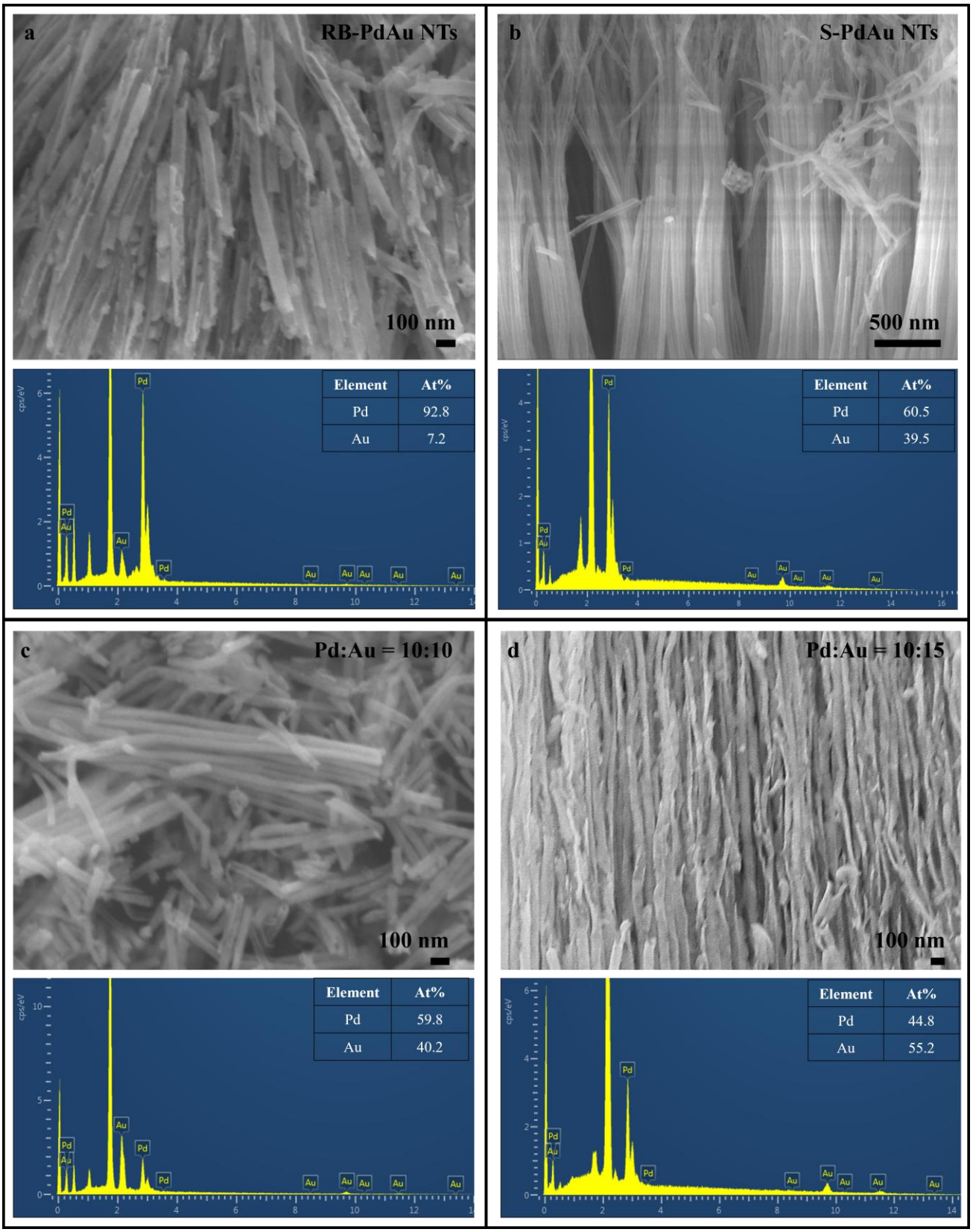

Fig. S2 The SEM and EDS analysis results of (a) RB-PdAu NTs, (b) S-PdAu NTs, (c) Pd:Au = 10:10 and (d) Pd:Au = 10:15.

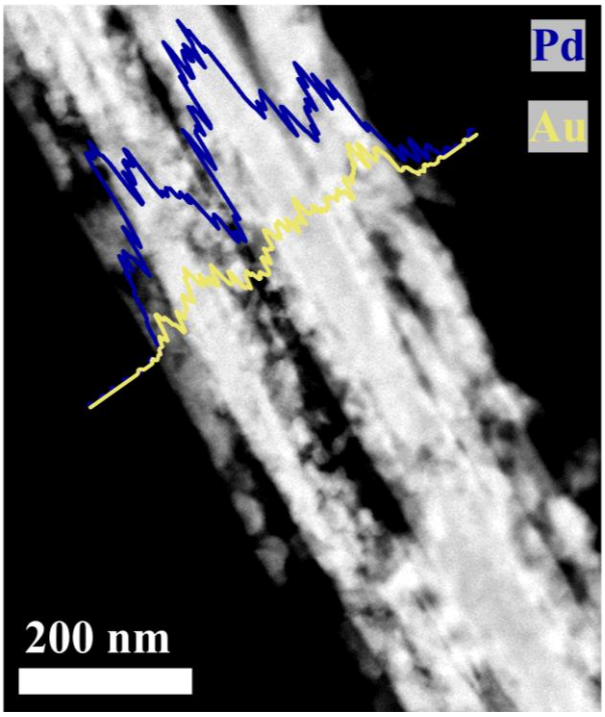

Fig. S3 The elemental line-scanning curves of Pd and Au on the RB-PdAu NTs.

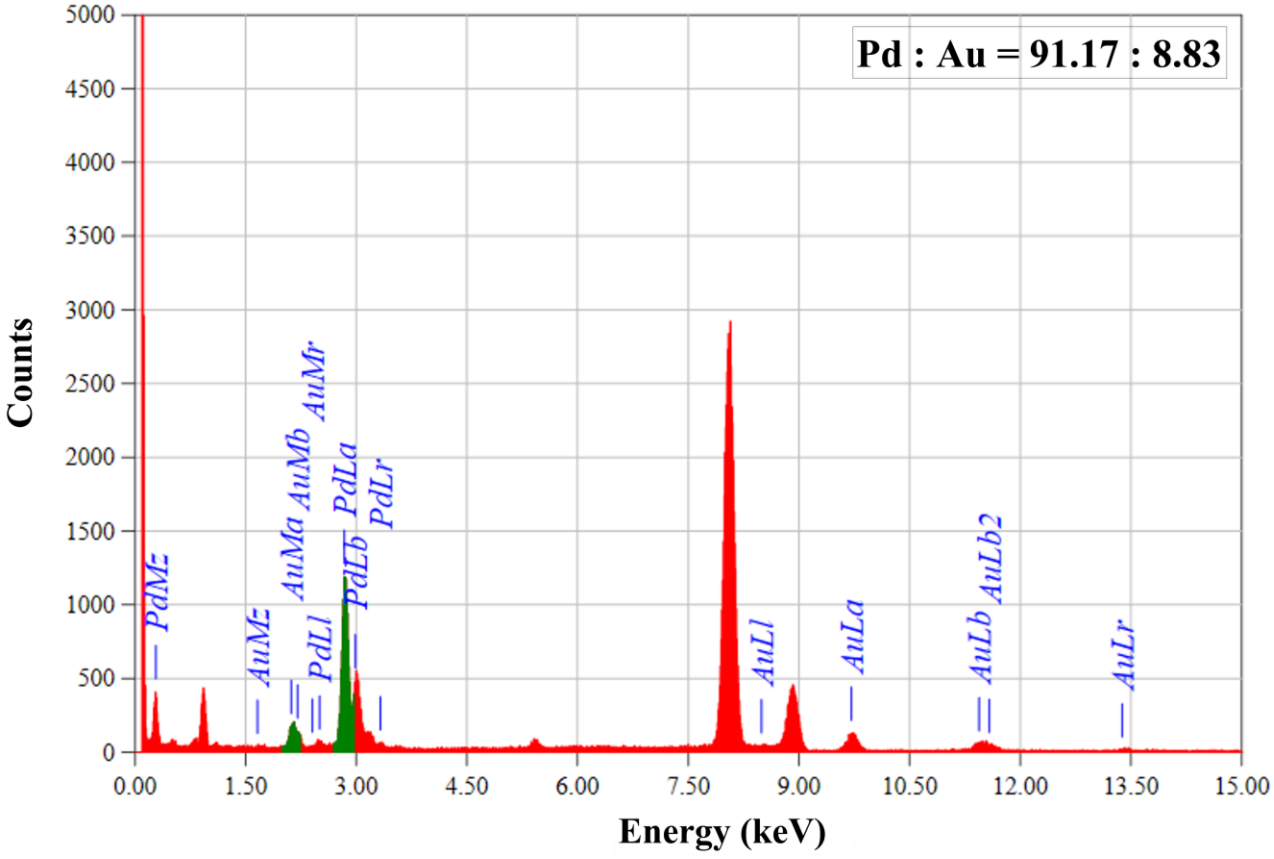

Fig. S4 The EDS pattern of RB-PdAu NTs.

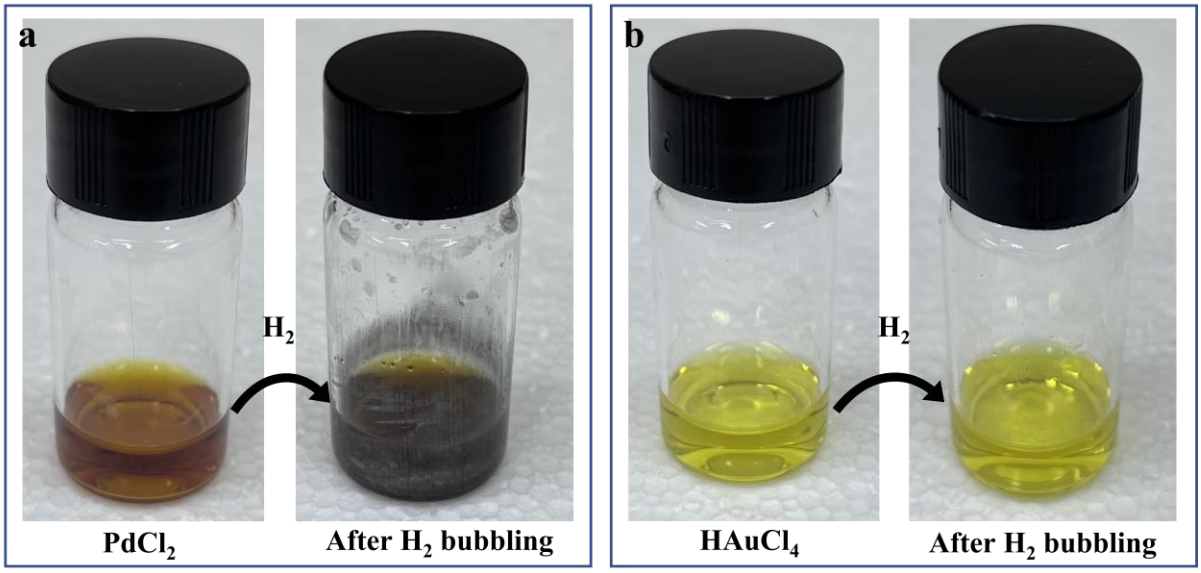

Fig. S5 The comparison for H_2 bubbling between (a) $PdCl_2$ and (b) $HAuCl_4$.

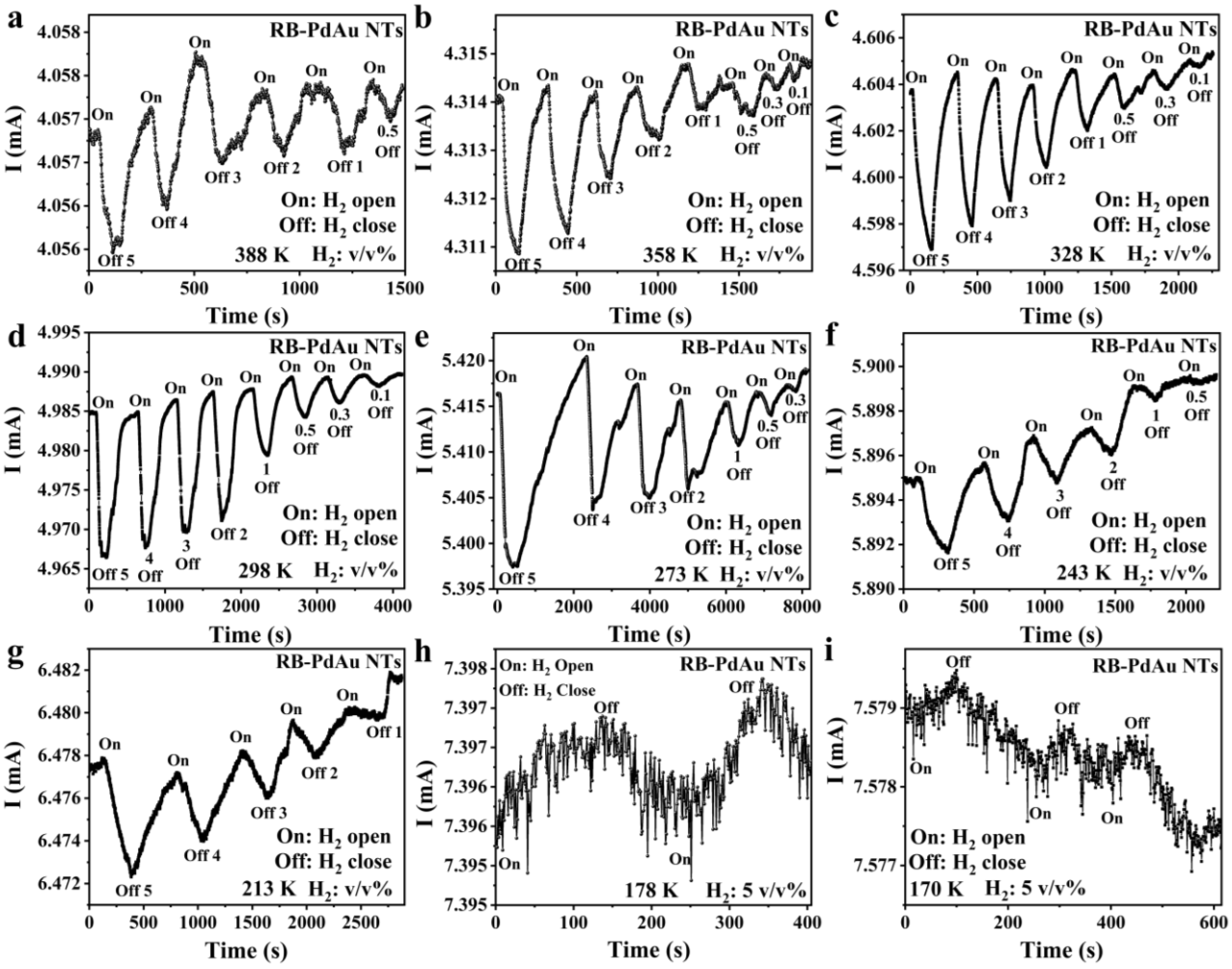

Fig. S6 The raw data of RB-PdAu NTs to various concentrations H_2 at various temperatures.

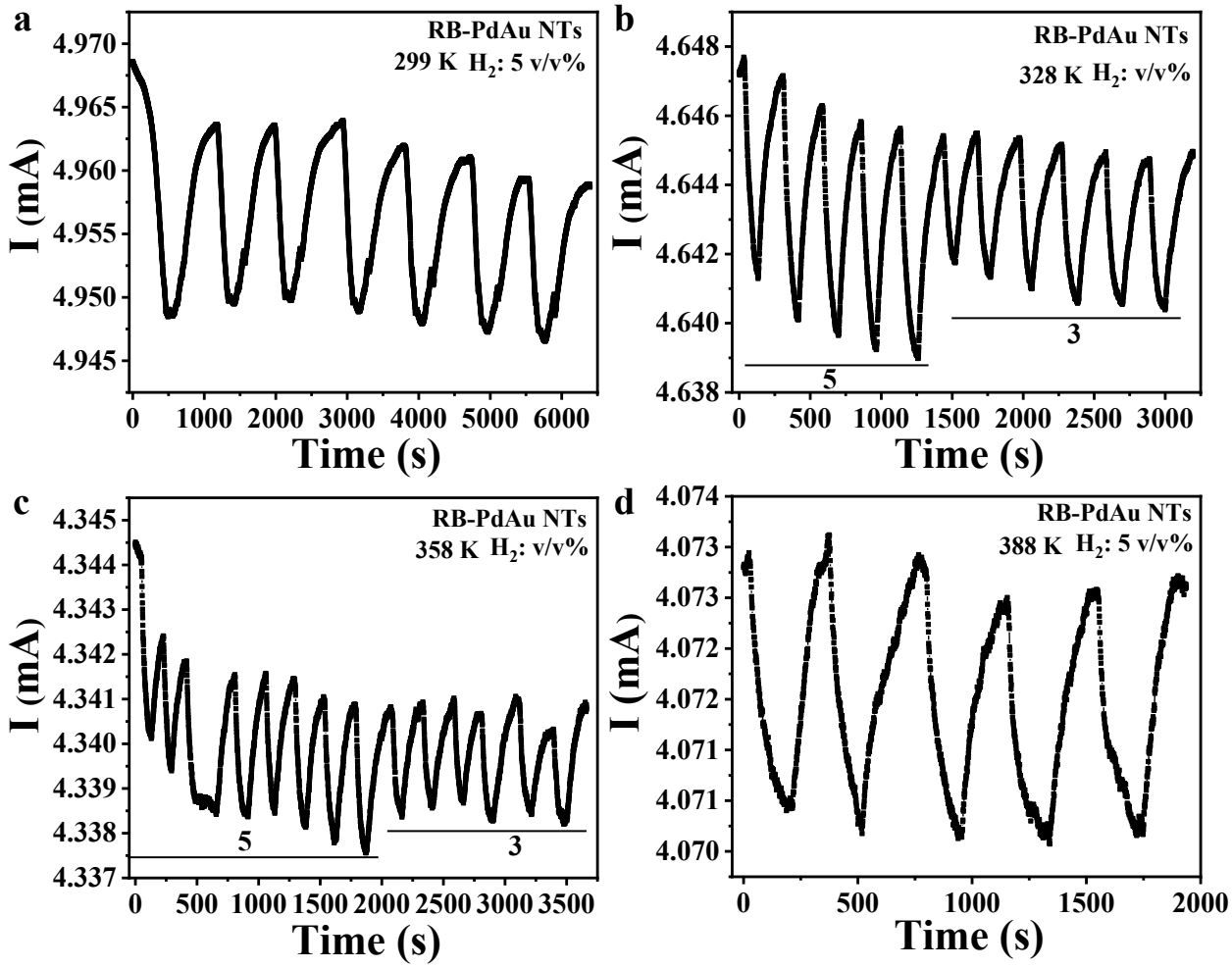

Fig. S7 The evaluation of RB-PdAu NTs after being exposed in low temperatures.

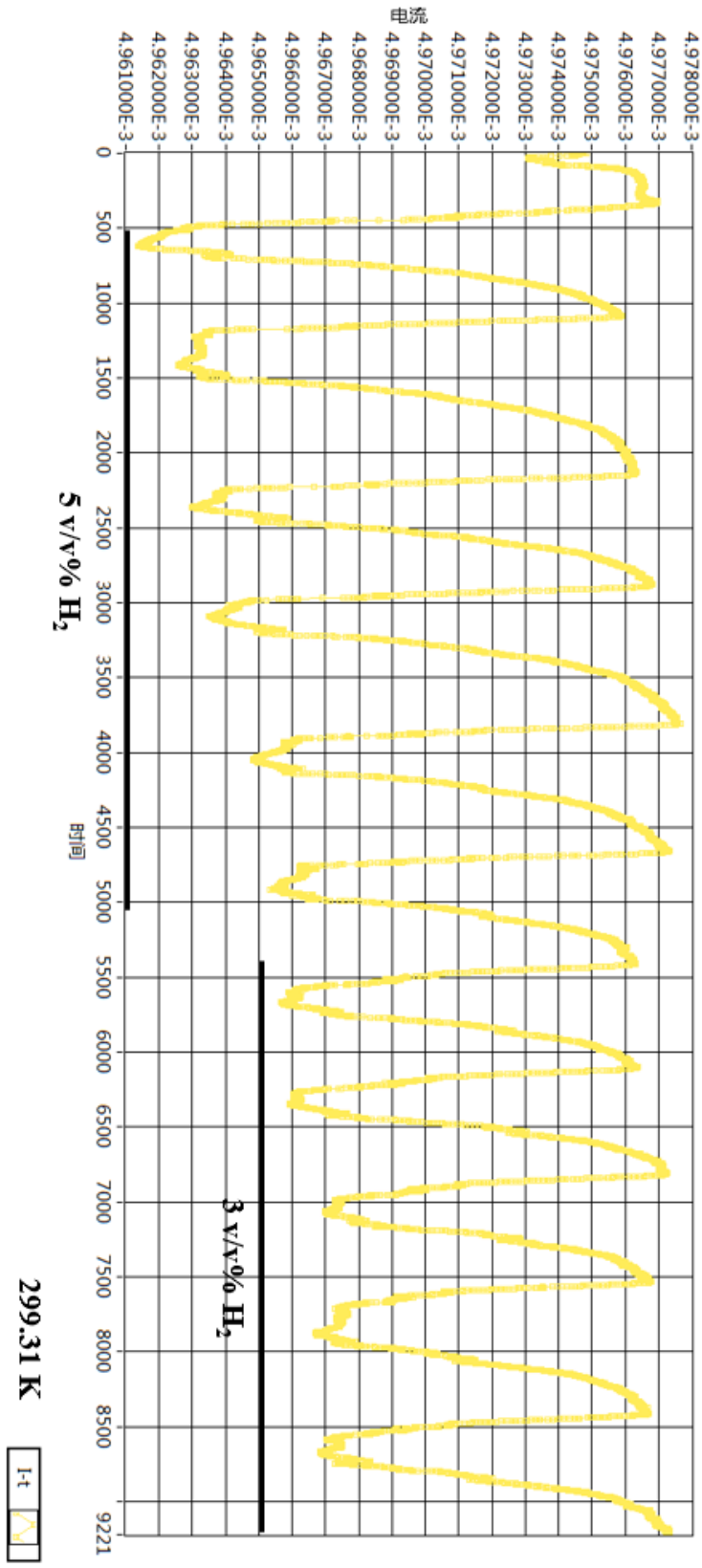

Fig. S8 A screenshot of RB-PdAu NTs for H_2 sensing at 299 K as an example.

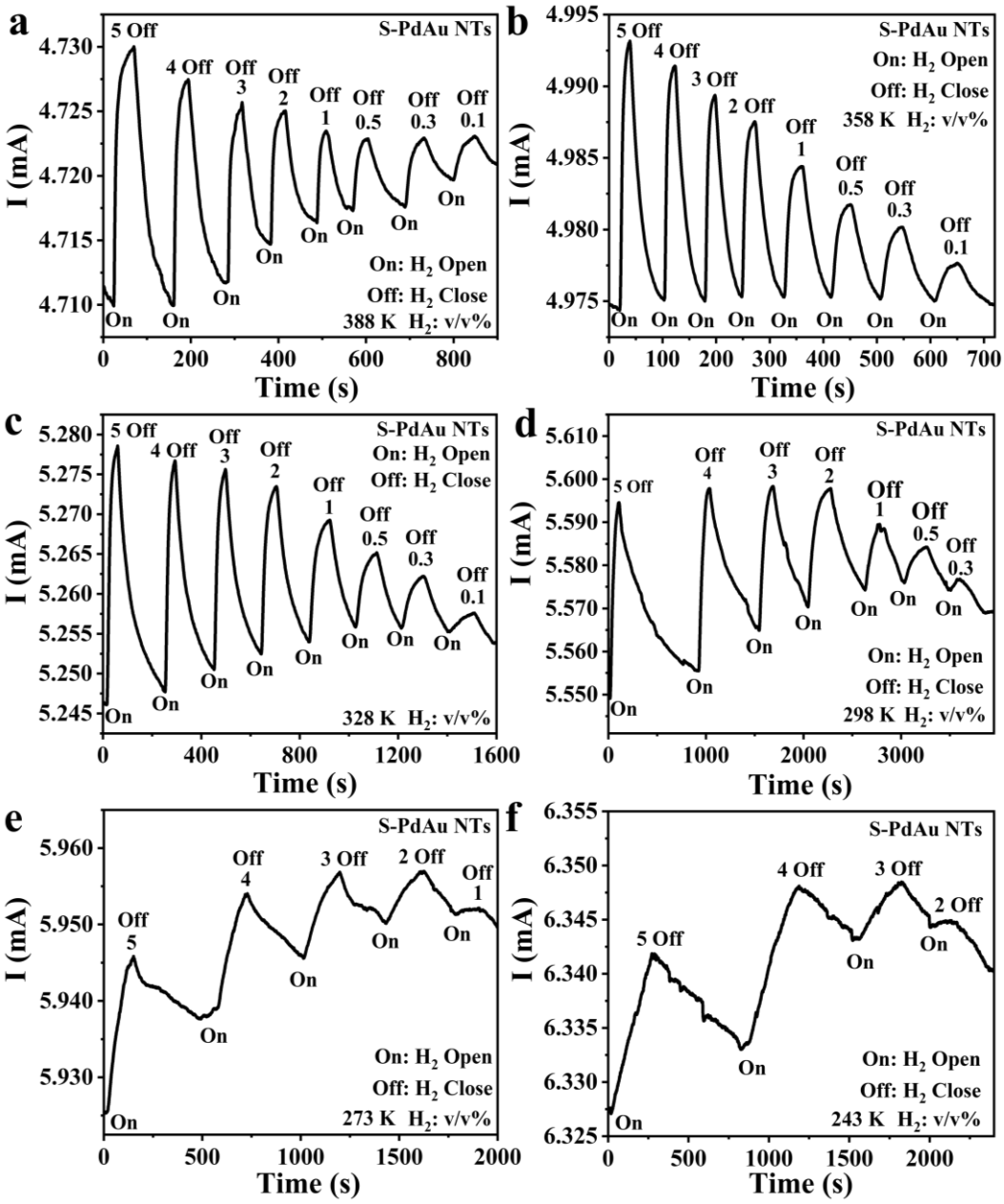

Fig. S9 The raw data of S-PdAu NTs for various concentrations H_2 at various temperatures.

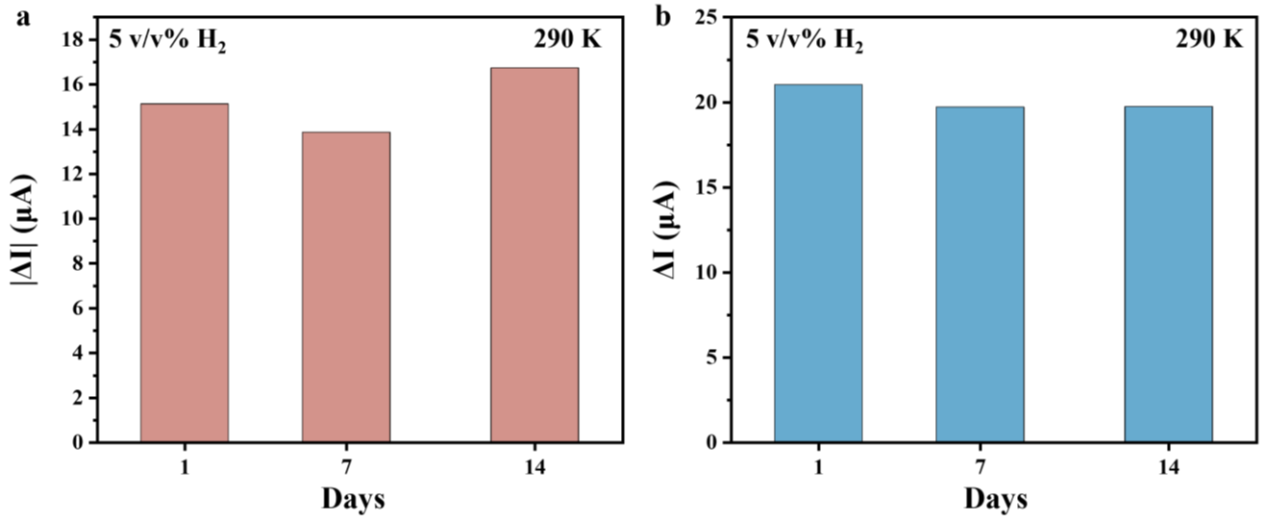

Fig. S10 The long-term stability test. (a) RB-PdAu NTs and (b) S-PdAu NTs.

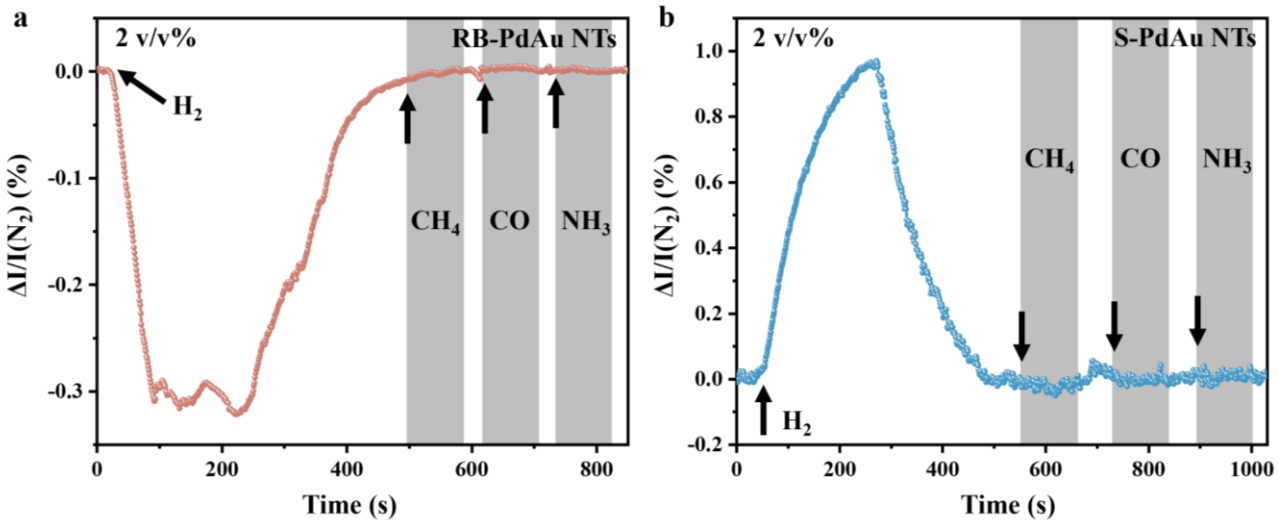

Fig. S11 The response curves of (a) RB-PdAu NTs and (b) S-PdAu NTs toward various gases.

Table S1 The quantitative comparison of response times for RB-PdAu NTs and S-PdAu NTs.

Temperature	388 K	358 K	328 K	298 K	273 K	243 K
Concentration	**Response time (s)** – RB-PdAu NTs / S-PdAu NTs					
5 v/v%	53 / 28	53 / 12	110 / 26	47 / 65	209 / 101	140 / 230
4 v/v%	40 / 22	78 / 12	71 / 26	51 / 75	142 / 119	108 / 311
3 v/v%	47 / 21	50 / 11	67 / 26	63 / 94	135 / 153	100 / 205
2 v/v%	43 / 19	46 / 13	65 / 27	88 /131	133 / 149	93 / 77
1 v/v%	50 / 10	32 / 17	53 / 51	109 / 115	145 / 85	78/ -

Table S2 Comparison of response time between our study to other existing low-temperature sensors.

Sensing Materials	Response time (s)			References
	3 v/v%	2 v/v%	1 v/v%	
Pure Pd NWs	~420	~220	~132	[1]
Pd/Bi/Cu HNAs	~147	~155	~109	[3]
RS-PdBi NWs	~80	~150	~300	[4]
zeolite-AFI@Pd NPs	~181	~204	~248	[5]
RB-PdAu NWs	**135**	**133**	**145**	**This study**
S-PdAu NWs	**153**	**149**	**85**	

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