## Supplementary material

- The schematic diagram of the commercial ceramic chip and the heater temperature and power vs. applied voltage curves; The schematic diagram of testing system for gas sensing.
- 2. SEM images of the samples: (a) S1, (b) S2, (c) S4 and (d) S5 at 50k.
- 3. Calculation process of the grain size.
- 4. Grain sizes and error bars of samples S1 S5.
- 5.  $N_2$  adsorption isotherms and BET surface area of the S2, S4 and S5 samples.
- 6. Pore size distributions of samples S1 S5.
- 7. Response and recovery curves of (a) S1; and (b) S3 to 200 ppm H<sub>2</sub> at 150 °C; (c) S1 to 200 ppm H<sub>2</sub> at 250 °C.
- 8. Response curves in stability of S1 and S3 sensors.
- 9. Theoretical limit of detection (LOD).
- 10. Current vs. Voltage behaviors of S1 and S3 sensors at 150 °C.
- 11. SEM images of the samples: (a) S1 and (b) S3 at 5k; (c) S3 at 50k.
- 12. Comparison of H<sub>2</sub> sensors based on different materials.

## High-response H<sub>2</sub> sensing performances of ZnO nanosheets

## modulated by oxygen vacancies

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Fig. S1 (a) The schematic diagram of the commercial ceramic chip; (b) The heater temperature and power vs. applied voltage curves; (c) The schematic diagram of testing system for gas sensing.



Fig. S2 SEM images of the samples: (a) S1, (b) S2, (c) S4 and (d) S5 at 50k.

Calculation process of the grain size.

The grain size is calculated according to the XRD results and Scherrer's equation:

$$D = \frac{K\lambda}{\beta \cos\theta}$$
(eqn S1)

where D is grain size, K is Scherrer constant (0.89),  $\lambda$  is CuK $\alpha$  wavelength (1.54056 Å),  $\beta$  is peak width at half-height (FWHM), and  $\theta$  is diffraction angle.

Here, we chose three main peaks ((100), (002), and (101)) to calculate the average grain size.

	Table. SI Grain sizes and error bars of samples SI – S5.					
Samples		Grain size (nm)	error bar (nm)			
-	<b>S</b> 1	16.58	4.11			
	S2	16.56	2.78			
	S3	15.99	2.79			
	S4	17.66	2.78			
	S5	15.64	3.17			



Fig. S3  $N_2$  adsorption isotherms and BET surface area of the S2, S4 and S5 samples.



Fig. S4 Pore size distributions of samples S1 – S5.



Fig. S5 Response and recovery curves of (a) S1; and (b) S3 to 200 ppm  $H_2$  at 150 °C; (c) S1 to 200 ppm  $H_2$  at 250 °C.



Fig. S6 Response curves in stability of S1 and S3 sensors.

Theoretical limit of detection (LOD).

According to the IUPAC (International Union of Pure and Applied Chemistry) definition, the signal could be considered as a true signal when the signal-to-noise ratio equals 3 [1]. Mathematically, the LOD could be calculated using the following equation  $LOD = 3 \frac{N_{rms}}{slope}$ (eqn S2)

where  $N_{rms}$  is the root Mean square (RMS) noise formula, and slope is extrapolated from the linear calibration curve as shown in Fig. 5c. The limit of detection here is calculated to be 55 ppb."



Fig. S7 Current vs. Voltage behaviors of S1 and S3 sensors at 150 °C.



Fig. S8 SEM images of the samples: (a) S1 and (b) S3 at 5k; (c) S3 at 50k.

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No.	Materials	Temp.	R	LOD	$T_{rec}$	Ref.			
		(°C)		(ppm)	(s)				
1	ZnO with defects	150	38.2	0.055	6	This			
						work			
2	SnO <sub>2</sub> with defect	250	2.21	0.1	12	[S2]			
3	ZnO/rGO composite	400	18	0.06	7	[S3]			
4	Pd-decorated	250	2.5	0.1	290	[S4]			
	ZnO nanosheet								
5	Pd-ZnO nanoflowers	RT	1.7	0.01	165	[S5]			
6	ZnO	190	1.25	0.01	960	[S6]			
7	ZnO@400 sensor	RT	2.15	5	6	[S7]			
8	Pd@ZnO-In <sub>2</sub> O <sub>3</sub>	350	42	5	240	[S8]			

Table. S2 Comparison of H<sub>2</sub> sensors based on different materials.

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