Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information: Electrical and Magnetic Properties of FeS_2 and $CuFeS_2$ Nanoplates

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1.Experiment

Synthesis of FeS_2 nanoplates: FeS_2 nanoplates were synthesized by a combination of hydrothermal method and annealing in CVD system . In a typical synthesis, 3.2 g of $FeSO_4{\cdot}7H_2O$ and 4 g of $Na_2S_2O_3$ were dissolved in 25 mL of de-ionized water under magnetic stirring. 0.7 g of S powder and 0.01 g of CTAB were added to the resulting solution under vigorous magnetic stirring at room temperature for 1 h. The formation reaction for FeS_2 can be summarized as follows:

$$Na_2S_2O_3 + S + FeSO_4 + H_2O \rightarrow Na_2SO_4 + FeS_2 + H_2SO_4 \quad (1)$$

Then this mixture was sealed in a teflonlined stainless steel autoclave (60% filled), maintained at 200 °C for 24 h, and cooled to room temperature naturally. Afterward, the black solid product was collected by centrifugation, washed with absolute alcohol, and dried naturally. Lastly, the sample was annealed in CVD system at 350 °C for 6 h, and would obtain about 1 g products.

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Synthesis of $CuFeS_2$ (FeS_2/$CuFeS_2$) nanoplates: 1.85 (2.5) g of $FeSO_4 \cdot 7H_2O$, 0.57 (0.24) g CuCl and 4 g of $Na_2S_2O_3$ were dissolved in 25 mL of de-ionized water under magnetic stirring. 0.7 g of S powder and 0.01 g of CTAB were added to the resulting solution under vigorous magnetic stirring at room temperature for 3 h. Then this mixture was sealed in a Teflonlined stainless steel autoclave (60% filled), maintained at 200 °C for 24 h, and cooled to room temperature naturally. Afterward, the black solid product was collected by centrifugation, washed with absolute alcohol, and dried naturally. Lastly, the sample was annealed in CVD system at 350 °C for 6 h.

Annealing in CVD system: A ceramic boat containing the sample was placed in the center of a tube furnace, another ceramic boat holding pure sulfur (S) was placed at the upwind low temperature zone in the quartz tube. During the reaction, the temperature of the low temperature zone was controlled to be a little above the melting point of S (113 °C). The quartz tube was first kept in a flowing protective atmosphere of high purity Ar (99.9999%) with the flow rate of 150-200 sccm. After 60 min of Ar purging, the furnace temperature was gradually increased from room temperature to 350 °C in 40 min. Then the temperature was kept at 350 °C for 6 h. The configuration was shown in Fig. S2.

Fabrication of FeS_2 and $CuFeS_2$ nanoplates devices: Firstly, the Au electrodes were fabricated with "gold-wire mask moving" technique: a micron gold-wire serving as a mask was fixed tightly on the surface of the silicon wafer, and then a pair of Au electrodes was deposited onto the substrate by thermal evaporation. After that, by slightly removing the Au wire mask, the Au electrodes were fabricated and a micron size gap was produced between the two electrodes. Then, FeS_2 or $CuFeS_2$ powder were dispersed in ethanol and dropped onto the surface of the gap between the two electrodes, and dried naturally at room temperature (Fig. S1).

DFT calculation: First-principles calculations for bulk FeS_2 and $CuFeS_2$ are performed on the basis of density functional theory using projector-augmented wave (PAW) potentials,[1] as implemented in the Vienna ab initio simulation package (VASP).[2] The generalized gradient approximation of Perdew-Burke-Ernzerhof (GGA-PBE) was adopted for exchange-correlation (XC) functional.[3] Energy cutoff for plane-wave expansion was set to 400 eV. The first Brillouin zone sam-

pling was performed with Monkhorst-Pack (MP) special k-point meshes.

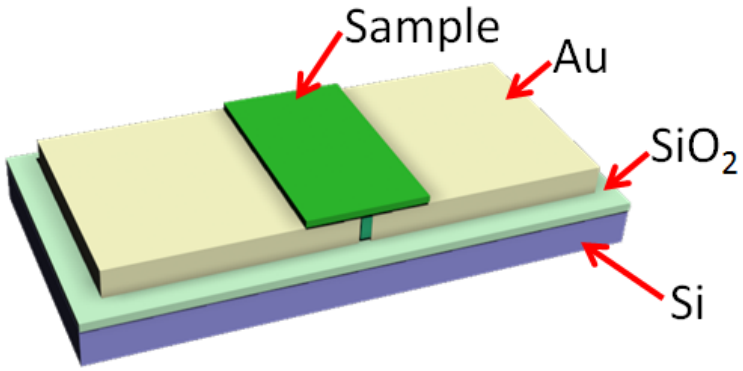

Fig. S1: The schematic view of the device.

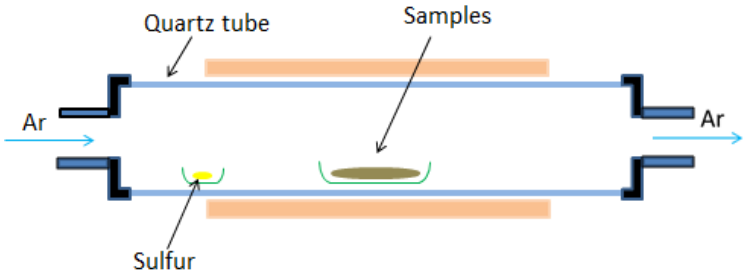

Fig. S2: The configuration used for annealing in our experiments.

2.Effects of annealing process and annealing temperatures on morphologies.

The annealing temperatures are 400 °C (Figure S3d), 450 °C (Figure S3e) and 500 °C (Fig. S3f) for FeS_2, respectively.

3.Analysis of $CuFeS_2$ SAED pattern.

There exist several sets of SAED patterns in the image. The red circles marked in the image (Fig. S4) showed a set of $CuFeS_2$ SAED pattern.

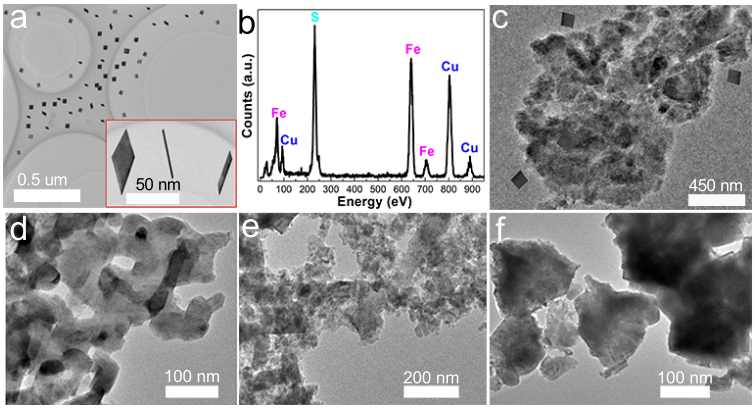

Fig. S3: LRTEM images of FeS_2 with annealing process at 350 °C (a) and without annealing process (c). (b), EDX-TEM profile of the FeS_2 nanoplates. (d), (e), (f) LRTEM images of FeS_2 at different annealing temperatures.

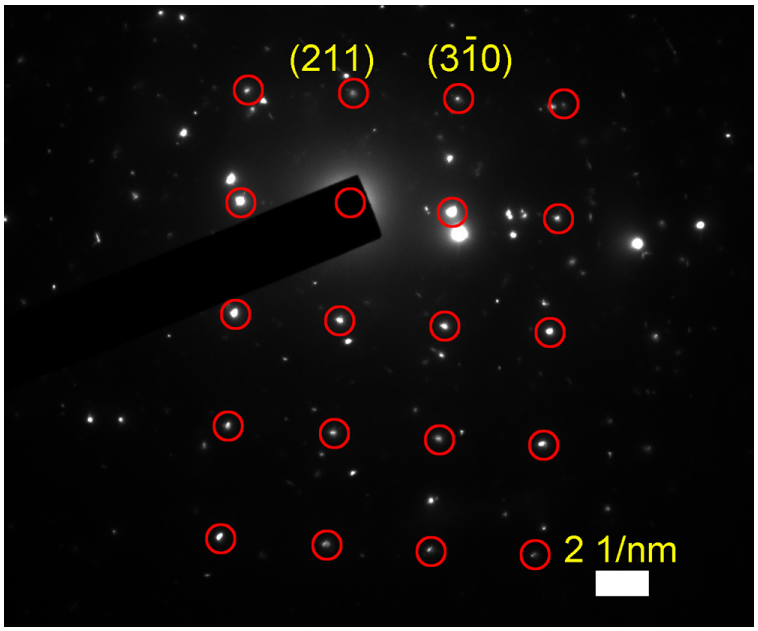

Fig. S4: Processed SAED pattern of $CuFeS_2$ nanoplates.

4.Raman spectra of FeS_2 and $CuFeS_2$ nanoplates.

The as-made products were measured by Raman spectra under 523 nm excitation (Fig. S5).

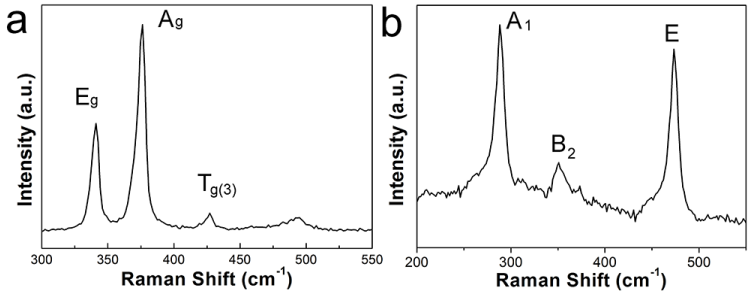

Fig. S5: Raman spectra of FeS_2 (a) and $CuFeS_2$ (b) nanoplates.

5.Comparison of XRD between FeS_2 and FeS_2/$CuFeS_2$ nanoplates in detail.

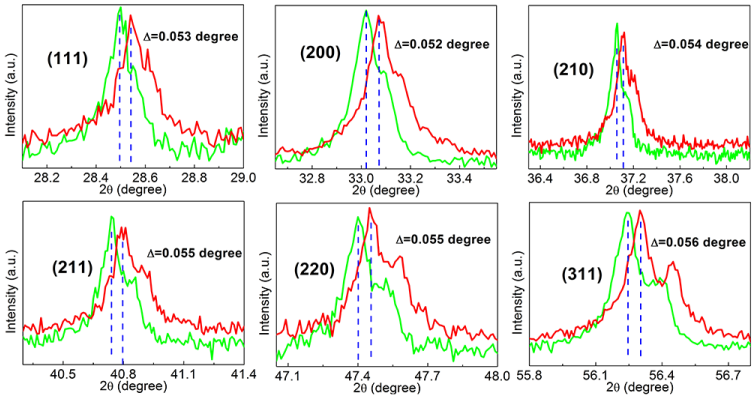

Fig. S6: Comparison of XRD peaks between FeS_2 and FeS_2/$CuFeS_2$ nanoplates at various positions. Red and green stood for XRD of FeS_2 and FeS_2/$CuFeS_2$ nanoplates, respectively. Δ represented difference values of XRD peak positions between FeS_2 and FeS_2/$CuFeS_2$.

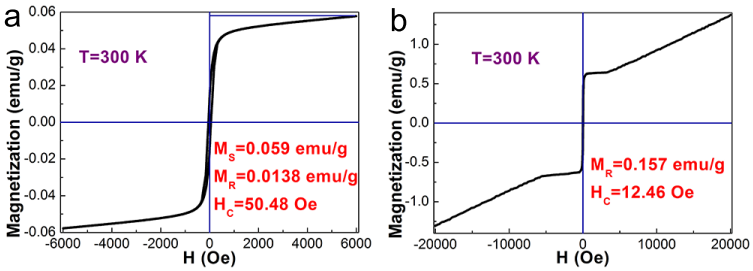

Fig. S7: Hysteresis loop of (a) 400 °C annealing and (b) 450 °C annealing FeS_2 at room temperature (300 K)

6.PPMS measurements of FeS_2 at different annealing temperatures.

References

(1) Blöchl, P. E. *Phys. Rev. B* **1994**, *50*, 17953–17979.

(2) Kresse, G.; Hafner, J. *Phys. Rev. B* **1993**, *47*, 558–561.

(3) Perdew, J. P.; Burke, K.; Ernzerhof, M. *Phys. Rev. Lett.* **1996**, *77*, 3865–3868.