Electronic Supplementary Information

Materials

All chemicals were of analytical grade and used directly without further purification. Rhodamine B, Fluorescein, Rhodamine 6G, 7-Hydroxycoumarin, hemoglobin, horseradish peroxidase, cytochrome c, catalase, glutathione reductase, lysozyme, acid fibroblast growth factor and basic fibroblast growth factor was purchased from Sigma-Aldrich. Sodium citrate and glucose oxidase were obtained from alfa aesar. Bovine serum albumin was achieved from Sangon (Shanghai, China). Copper nitrate trihydrate, sodium sulfide, other reagents and solvents were achieved from Beijing Chemicals (Beijing, China).

Synthesis of sodium citrate-templated CuS nanoparticle

In the experiment, sodium citrate-templated CuS nanoparticles were synthesized by a modified method reported previously.[33]The general procedure for the synthesis of CuS NPs in water was as follows. Into 250 mL of an aqueous solution containing $Cu(NO_3)_2$ (0.25 mmol) and sodium citrate (0.17 mmol) was added 250 μL of sodium sulfide solution (Na_2S, 1 M) with stirring at room temperature. The pale-blue $Cu(NO_3)_2$ solution turned dark-brown immediately upon the addition of sodium sulfide. After stirring for 5 min, the reaction mixture was heated to 90 °C and stirred for 15 min until a dark-green solution was obtained. The mixture was transferred to ice-cold water. The citrate-templated CuS NPs (Cit-CuS NPs) were obtained. The obtained particles were centrifuged and washed with water and stored in a PBS buffer in 4 °C.

Fluorescence quenching and recovery experiments

In the fluorescence quenching experiments, 3 samples of each fluorescent dyes solutions were prepared in diluted water. After adding 300 μL CuS solutions (3 mg/mL), the mixtures were incubated in 20 °C for 10 min. The control group added PBS buffer instead of CuS solutions. Fluorescent spectra were then measured individually by excitation at 550 nm, 495 nm, 525 nm, 320 nm, respectively. In the fluorescence recovery experiments, the mixtures of CuS and fluorescent dyes were incubated with BSA solution (100 μL 1 mM) in 20 °C for 30 min while dilute water was added to the mixture for control. Fluorescent spectra were then measured individually.

Preparation of the bacteria extract

MRSA (ATCC29213) and MSSA (ATCC25923) were grown in LB medium at 37 °C. The bacterial cells were isolated by centrifugation (3500 rpm). The bacterial cells were rinsed with PBS buffer (50 mM, pH 7.4), and designed bacterial concentration was adjusted by measuring the optical density at 600 nm (OD_{600nm}). The bacteria were dispersed in PBS buffer and lysed under ultrasonic (600 W) for 3 hours. After centrifuged and removed the precipitation, the bacteria extracts were stored in 4 °C

Sensing proteins and bacteria extracts.

The dyes-CuS probes containing 600 μL fluorescent dyes and 300 μL CuS were prepared by incubated in 20 °C in a PBS buffer. The 10 proteins were dispersed in diluted water to obtained 1 mM solutions. 100 μL of each protein solution were mixed with 4 kinds of dyes-CuS probes individually and incubated in 20 °C for 30 min. The

bacteria extracts (100 μL, 0.5 OD) were incubated with the dyes-CuS probes in 20 °C for 30 min. Fluorescence of 5 replicates were measured for each protein-dye or extracts combinations. The control group used PBS buffer instead of protein solutions or bacteria extracts.

Sensing array on quartz chips.

10 quartz chips were pretreated by ultrasonic in ethanol and sodium dodecyl sulfate solution for 30 min, respectively. The chips were washed with water and air-dried before used. 5 μL of 4 kinds of dye-CuS probes were dropped on different position of the chips in proper sequence followed by dried in room temperature for 30min. Protein samples (5 μL) were dropped on the corresponding location of probes and incubated for 5 min. After that, the probe chips were moved onto a UV transilluminator and the images were taken directly by a digital camera.

PCA

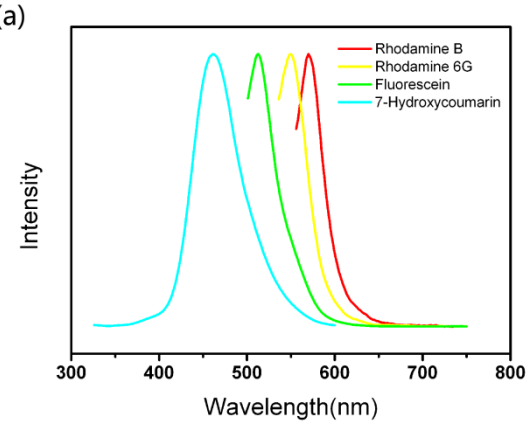
The PCA was carried out by using MATLAB R2012a and Origin 8.5. The code of PCA program was shown as follows:

```
clear;
load F:\workspace\RB.txt;
load F:\workspace\FL.txt;
load F:\workspace\R6G.txt;
load F:\workspace\C.txt;
load F:\workspace\N.txt;

for i=1:N
    for j=1:5
        b((i-1)*5+j,1)=RB(i,j);
        b((i-1)*5+j,2)=FL(i,j);
        b((i-1)*5+j,3)=R6G(i,j);
        b((i-1)*5+j,4)=C(i,j);
    end
```

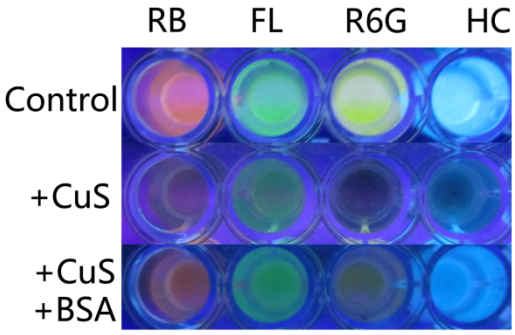
```
end

cor=corrcoef(zscore(b));
[d,v]=eig(cor);
disp (v);
disp (b);
ou=b*d;
disp (ou);

save F:\workspace\V.txt v -ascii;
save F:\workspace\training.txt b -ascii;
save F:\workspace\PCA.txt ou -ascii;
```

The output files were analyzed by utilizing Origin 8.5.

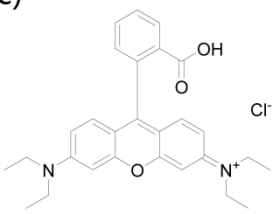
(a)



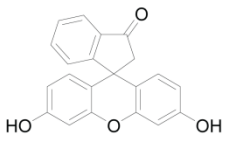
(b)



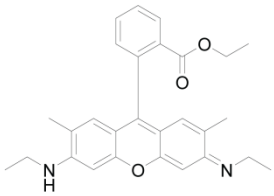
(c)



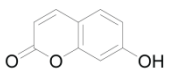
Rhodamine B



Fluorescein



Rhodamine 6G



7-Hydroxycoumarin

Fig. 1 (a) The fluorescence spectra of Rhodamine B, Fluorescein , Rhodamine 6G, 7-Hydroxycoumarin (b) The dye sensor array treated without CuS, with CuS and with both CuS and BSA under the irradiation of UV lamp (c) The chemical structures of 4 dyes used in the study.

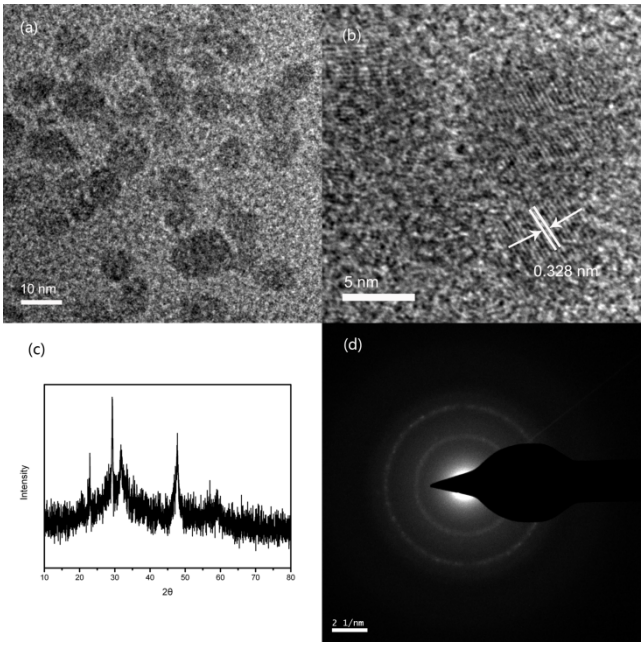


Fig. S2 (a) Low-magnification and (b) high-magnification TEM images of the CuS NPs. (c) Powder XRD patterns of the CuS NPs. (d) Selected area electron diffraction (SAED) pattern of the CuS NPs

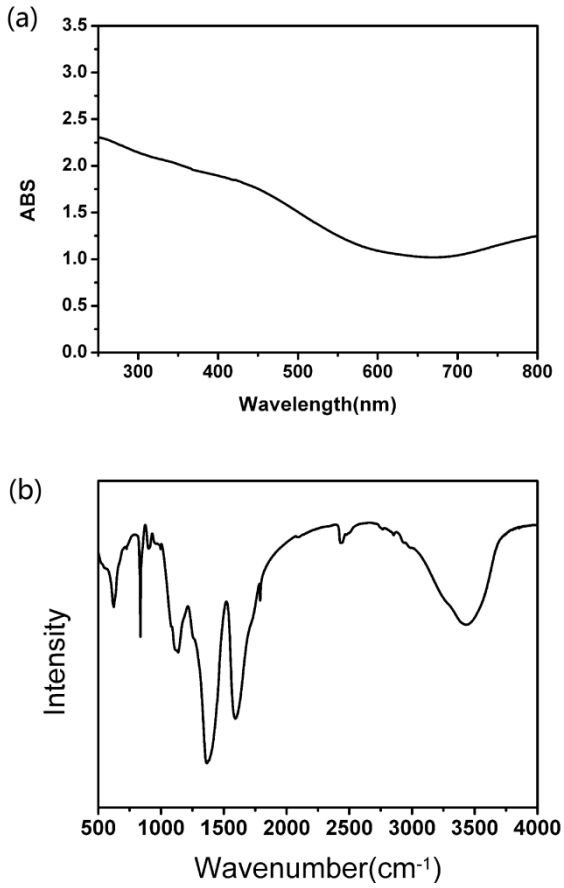


Fig. S3 (a) UV-Vis-NIR spectroscopy of the CuS aqueous solution. (b) The FTIR spectra of the sodium citrate templated CuS nanoparticles.

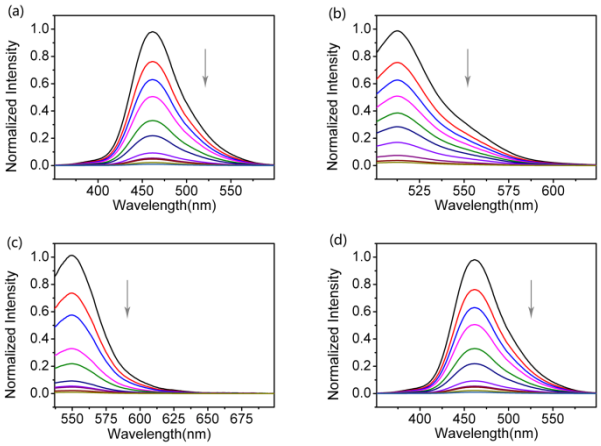


Fig. S4 The fluorescent spectra of (a) Rhodamine B (b) Fluorescein (c) Rhodamine 6G and (d) 7-Hydroxycoumarin upon adding different concentration of CuS. The fluorscent intensity declined as the concentration of CuS enhanced (concentration of CuS:0-30 mg/L for Rhodamine B, 0-100 mg/L for Fluorescein and Rhodamine 6G, 0-200 mg/L for 7-Hydroxycoumarin).

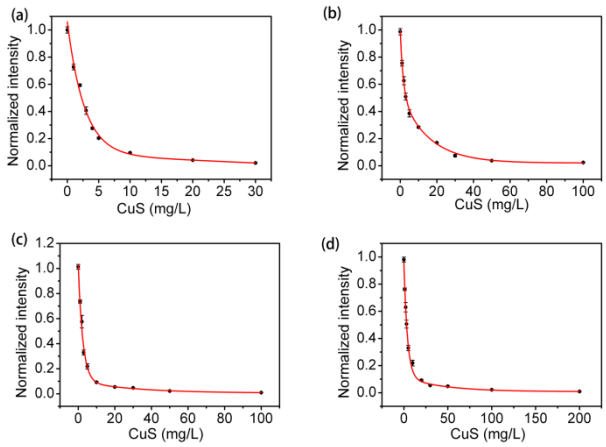


Fig. S5 Fluorescence-quenching curves of (a) Rhodamine B (b) Fluorescein (c) Rhodamine 6G and (d) 7-Hydroxycoumarin used in the initial tongue array (concentration of CuS: 0-30 mg/L for Rhodamine B, 0-100 mg/L for Fluorescein and Rhodamine 6G, 0-200 mg/L for 7-Hydroxycoumarin).

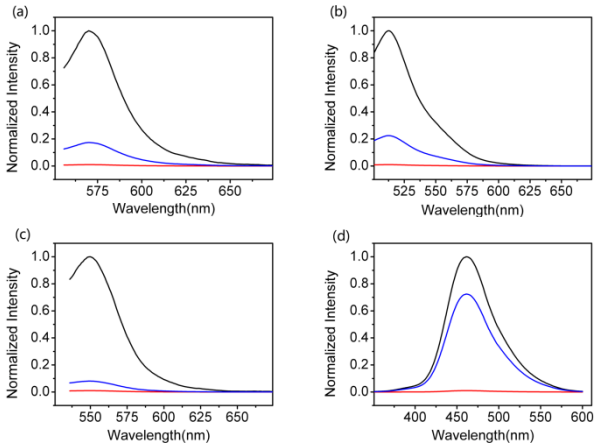


Fig. S6. The fluorescent spectra of (a) Rhodamine B (b) Fluorescein (c) Rhodamine 6G and (d) 7-Hydroxycoumarin add nothing (black line), CuS (red line) and CuS followed by BSA (blue line).

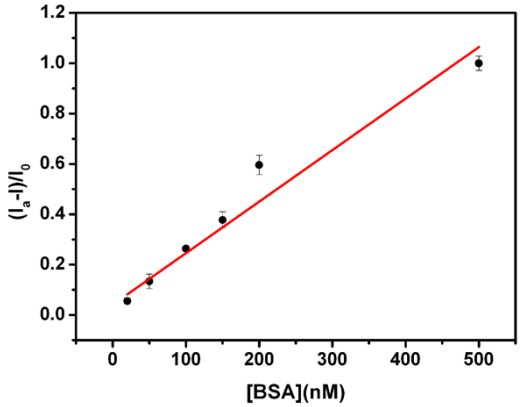


Fig. S7 The linear region of the $(I_a\text{-}I)/I_0$-[BSA] curve.

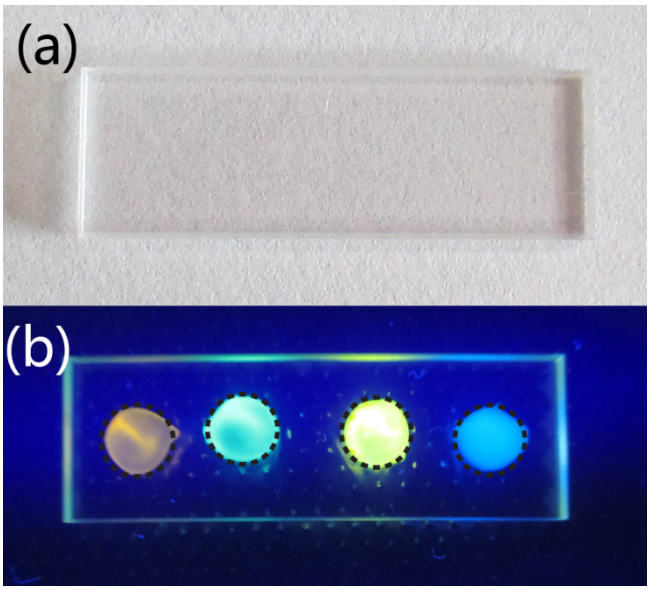


Fig. S8 (a) Photo of the quartz chip with pretreated by ultrasonic in ethanol and sodium dodecyl sulfate solution. (b) Photo of fluorescence of 4 dyes on the chip under UV irradiation.

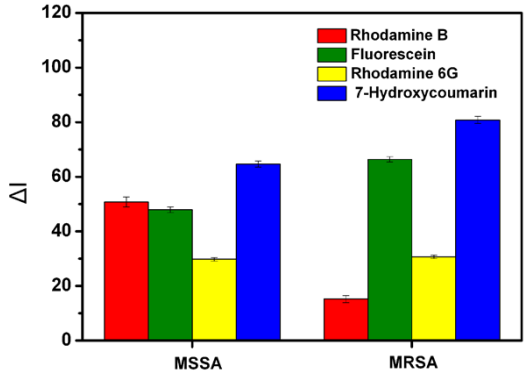


Fig. S9. Fluorescence response (ΔI) of the CuS array in presence of the extracts of MSSA and MRSA.

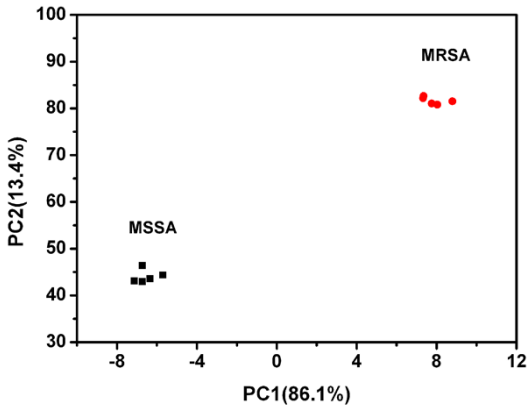


Fig. S10 The PCA score plots of extracts of MSSA and MRSA measured five times.

Table S1. The molecular weights and the isoelectric points of the 10 proteins.

	M.W.	I.P.
BSA	66 kDa	4.7
Hb	67 kDa	7.2
HRP	40 kDa	7.2
Cc	12 kDa	10.7
Gox	16 kDa	4.6
CAT	232 kDa	4.7
GR	56 kDa	6.75
LYSO	14 kDa	11.35
aFGF	16 kDa	5.6
bFGF	18 kDa	9.6

Table S2. The identifying results of 30 unknown protein samples.

Label	RB	FL	R6G	HC	Identity	Prediction accuracy
1	261.8	383.3	375.25	141.33	LYSO	Y
2	54.78	365.2	7.36	79.09	GOx	Y
3	87.13	159.19	59.58	301.18	BSA	N
4	45.3	178.22	26.6	-17.58	Cc	Y
5	211.52	383.37	231.9	183.94	GR	Y
6	77.02	124.85	63.1	282.04	Hb	Y
7	51.19	243.22	84.87	182.9	bFGF	Y
8	80.56	380.02	52.06	238.23	aFGF	Y
9	51.78	364.56	8.02	76.28	GOx	Y
10	55.18	80.26	26.49	345.39	HRP	Y
11	45.13	148.54	23.7	-17.36	Cc	N
12	213.49	381.25	232.67	179.95	GR	Y
13	85.93	161.43	40.38	302.75	BSA	Y
14	261.4	385.14	377.57	139.16	LYSO	Y
15	174.78	405.04	63.98	85.03	CAT	Y
16	42.38	177.58	24.64	-16.95	Cc	Y
17	179.01	384.65	202.46	184.95	GR	N
18	50.18	232.75	83.87	185.37	bFGF	Y
19	78.55	125.15	60.65	283.49	Hb	Y
20	54.99	79.7	26.54	350.01	HRP	Y
21	80.04	383.62	55.99	235.63	aFGF	Y
22	195.35	384.11	62.45	80.68	CAT	N
23	84.41	390.2	55.29	235.73	aFGF	Y
24	171.23	408.82	61.92	83.03	CAT	Y
25	55.76	364.2	7.7	76.46	GOx	Y
26	78.09	125.58	60.78	281.3	Hb	Y
27	50.18	242.55	85.1	182.67	bFGF	Y
28	55.59	80.29	26.29	350.01	HRP	Y
29	262.8	384.3	377.25	139.33	LYSO	Y
30	88.4	157.89	41.58	304.86	BSA	Y