

# Supporting Information

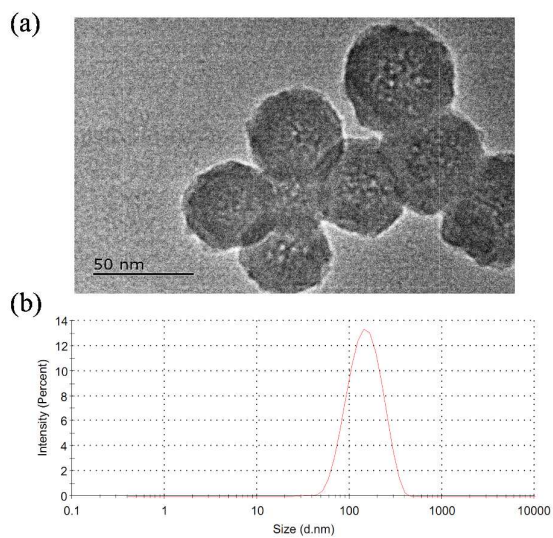
## A Dual-Ratiometric Fluorescent Nanoprobe for Visualizing the Dynamic Process of pH and Superoxide Anion Changes in Autophagy and Apoptosis

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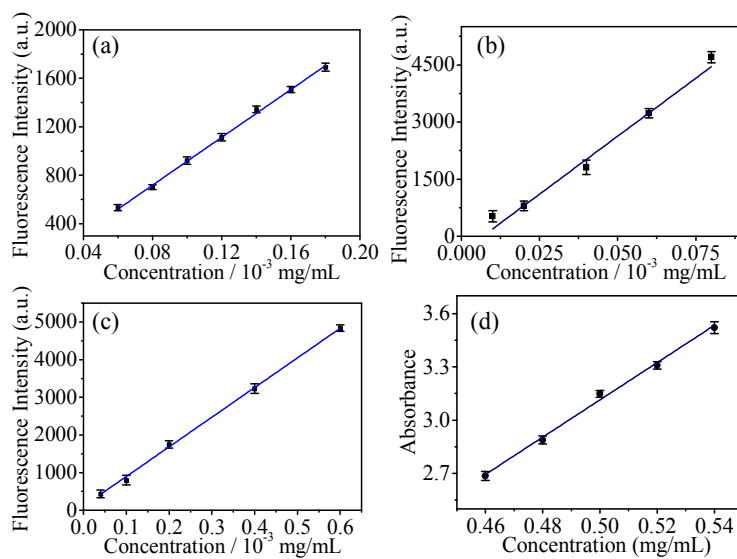
College of Chemistry, Chemical Engineering and Materials Science, Collaborative Innovation Center of Functionalized Probes for Chemical Imaging in Universities of Shandong, Key Laboratory of Molecular and Nano Probes, Ministry of Education, Institute of Molecular and Nano Science, Shandong Normal University, Jinan 250014, (P. R. China).

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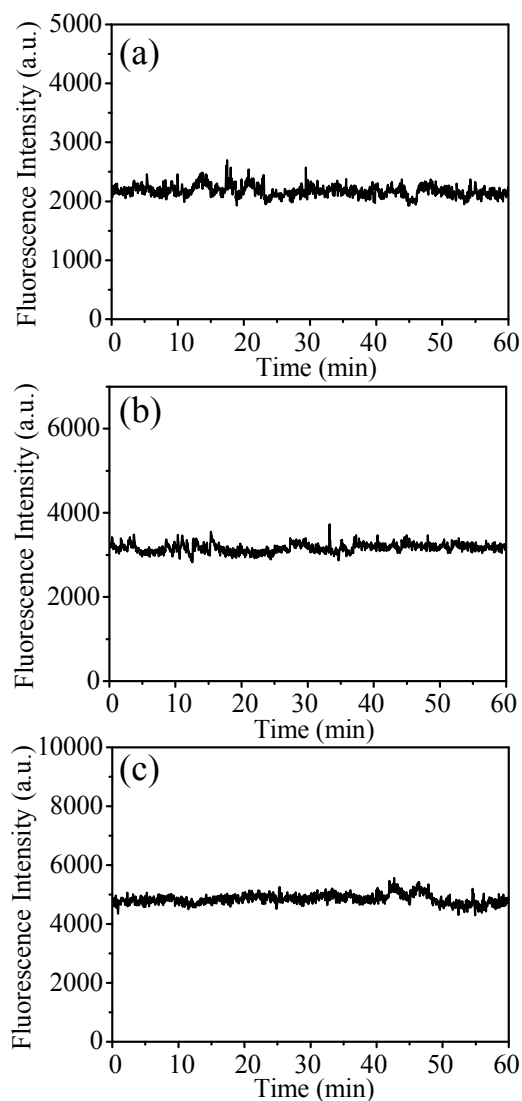
**Supplementary Figure:**



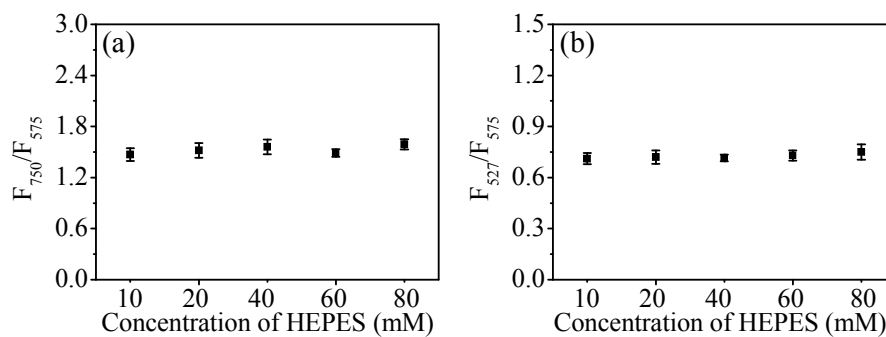
**Figure S1.** TEM image (a) and dynamic light scattering histograms (b) of the nanoprobe.



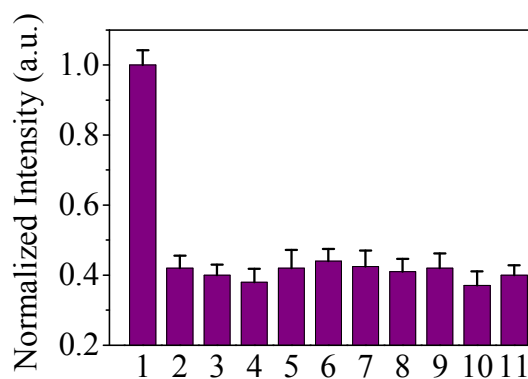
**Figure S2.** Standard linear calibration curves of RhB (a), DBZTC (b), Tpy-Cy (c) and TPP (d).



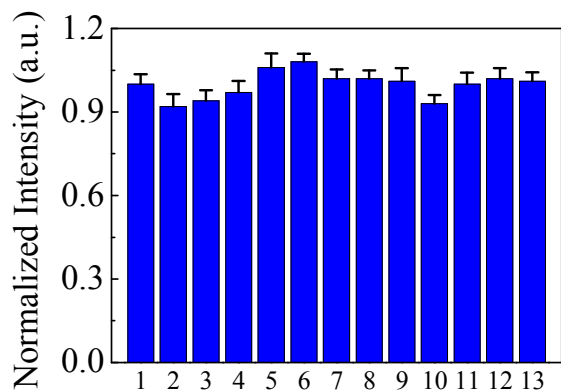
**Figure S3.** The stability of the nanoprobe under continuous light illumination. (a) The fluorescence kinetic of DBZTC. The excitation and emission wavelength were 470 nm and 527 nm, respectively. (b) The fluorescence kinetic of RhB. The excitation and emission wavelength were 560 nm and 575 nm, respectively. (c) The fluorescence kinetic of Tpy-Cy. The excitation and emission wavelength were 650 nm and 750 nm, respectively.



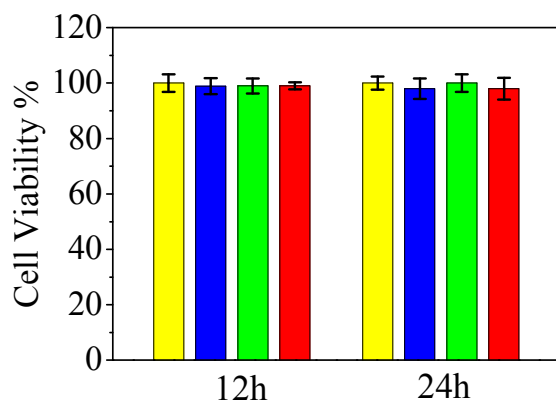
**Figure S4.** The fluorescence intensity ratio  $F_{527}/F_{575}$  (a) and  $F_{750}/F_{575}$  (b) in different concentration of HEPES.



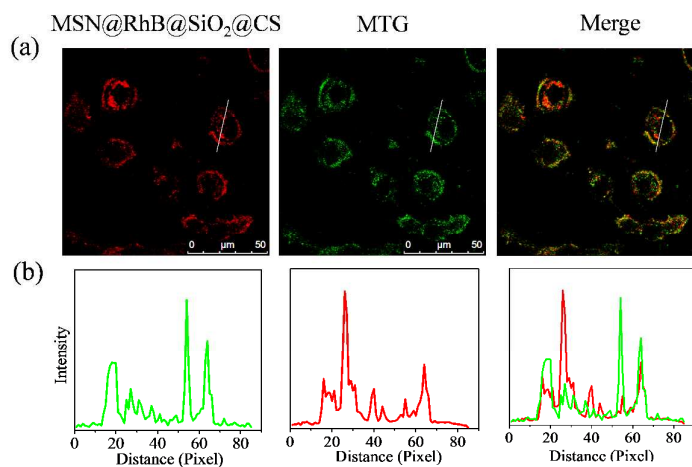
**Figure S5.** Fluorescence response of nanoprobe in the presence of different species. 1:  $O_2^{\cdot-}$  (1.7  $\mu M$ ); 2:  $^1O_2$  (0.35 mM); 3:  $ClO^-$  (1.7 mM); 4: GSH (0.35 mM); 5:  $H_2O_2$  (1.7 mM); 6: NO (17.0  $\mu M$ ); 7:  $\cdot OH$  (17  $\mu M$ ); 8:  $ONOO^-$  (3.5  $\mu M$ ); 9: t-BuOOH (0.35 mM); 10: VC (0.17 mM); 11: control. The concentration of the nanoprobe was 0.2 mg/mL.



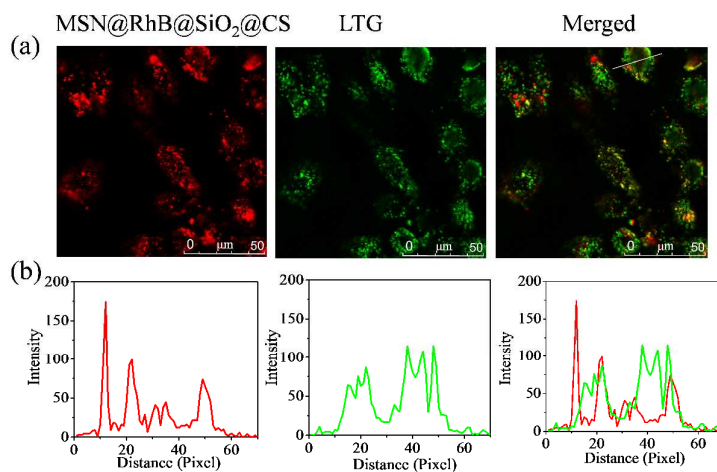
**Figure S6.** Fluorescence response of nanoprobe in the presence of intracellular species. 1: blank; 2: Cu<sup>2+</sup> (0.3 mM); 3: Fe<sup>2+</sup> (0.3 mM); 4: Mn<sup>2+</sup> (0.3 mM); 5: Zn<sup>2+</sup> (0.3 mM); 6: Co<sup>2+</sup> (0.3 mM); 7: Ca<sup>2+</sup> (0.5 mM); 8: Mg<sup>2+</sup> (0.5 mM); 9: H<sub>2</sub>O<sub>2</sub> (1.0 mM); 10: GSH (0.35 mM) 11: K<sup>+</sup> (10.0 mM); 12: Na<sup>+</sup> (10.0 mM). The concentration of the nanoprobe was 0.2 mg/mL.



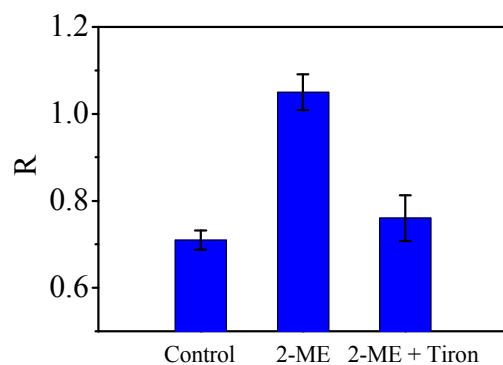
**Figure S7.** MTT assay of HeLa cells incubated with nanoprobe for 12 h and 24 h. Yellow bars stand for the control, blue bars, green bars and red bars stand for the nanoprobe 0.2, 0.4 and 0.8 mg/mL, respectively.



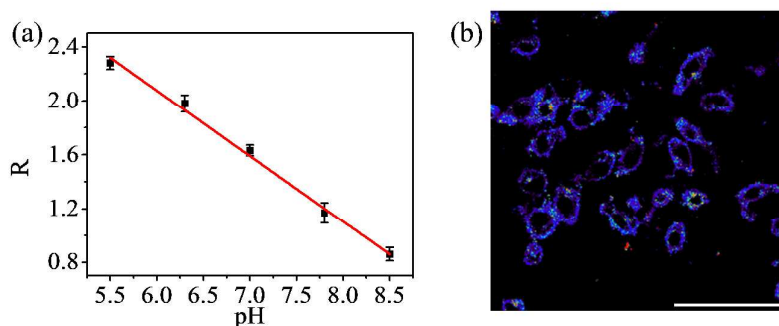
**Figure S8.** (a) Confocal fluorescence images of HeLa cells incubated with MSN@RhB@SiO<sub>2</sub>@CS and MTG. (b) Line-scan profiles of fluorescence intensity in the corresponding confocal images in a.



**Figure S9.** (a) Confocal fluorescence images of HeLa cells incubated with MSN@RhB@SiO<sub>2</sub>@CS and LTG. (b) Line-scan profiles of fluorescence intensity in the corresponding confocal images in a.

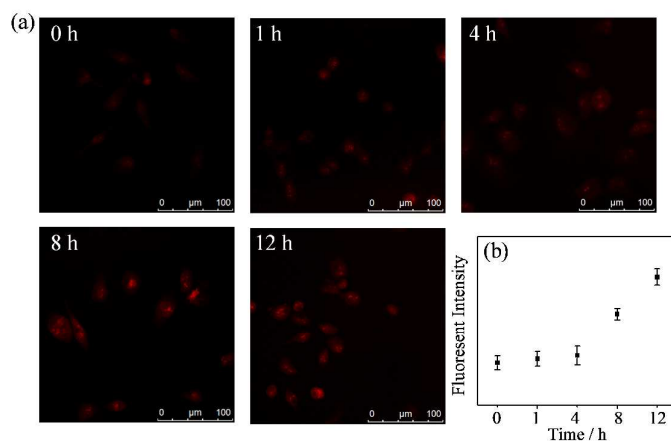


**Figure S10.** Average  $F_{\text{green}}/F_{\text{Purple}}$  intensity ratios (R) for the ratio images in Figure 4.



**Figure S11.** Intracellular pH calibration curve of nanoprobe in HeLa cells. (b)

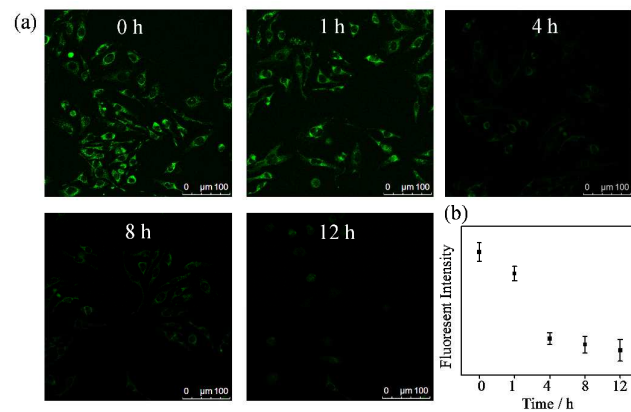
Ratiometric imaging of intact cells. Scale bars: 100  $\mu\text{m}$ .



**Figure S12.** Fluorescence imaging of HeLa cells stained by acridine orange after

treated by GP-starvation (a) for 0, 1, 4, 8 and 12 h. (b) AVOs were represented by the

red puncta dots in the fluorescent pictures. Scale bars: 100  $\mu\text{m}$ .



**Figure S13.** (a) Fluorescence imaging of HeLa cells loaded with rhodamine 123 during BSO-induced apoptosis for various times (0-12 h). (b) Mean fluorescence intensity of the nanoprobe treated cells in (a).