Supplementary Data

NIR Remote-controlled "Lock-Unlock" Nanosystem for Imaging Potassium Ions in Living Cells

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Table S1. Sequences of oligonucleotides used in this work.

Name	Sequence (5'-3')
A1	Texas-Red-GAGTATTTCTCTACGGGTTAGGGTTAGGGTTAGGGT-Cy3 ↓ K^+ aptamer
three-base mismatched strand A2	Texas-Red-GAGTATTTCTCTACGGGTTAGCGTTAGAGTTAGCGT-Cy3
C1	CTAACCCGTAGAGAAATACTCT$_{(10)}$-SH

Supporting Figures

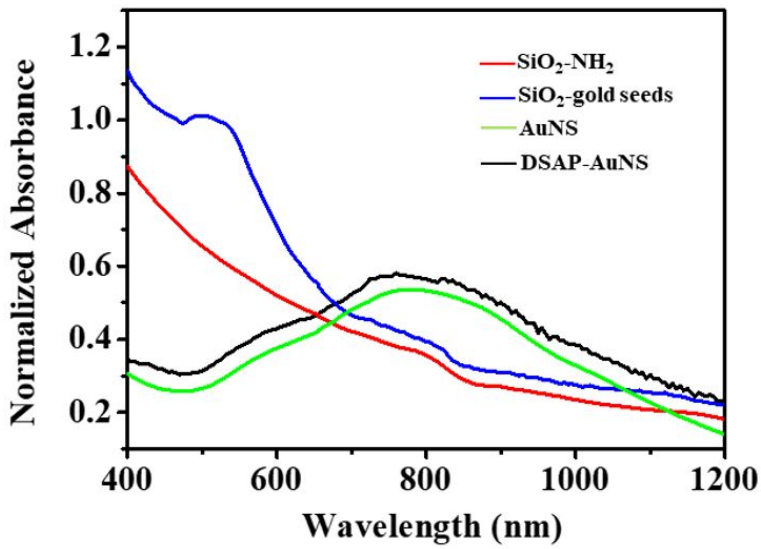

Figure S1. UV-*vis* absorption spectra of SiO_2-NH_2 (red), SiO_2-gold seeds (blue), AuNS (green) and DSAP-AuNS (black) in buffer solution.

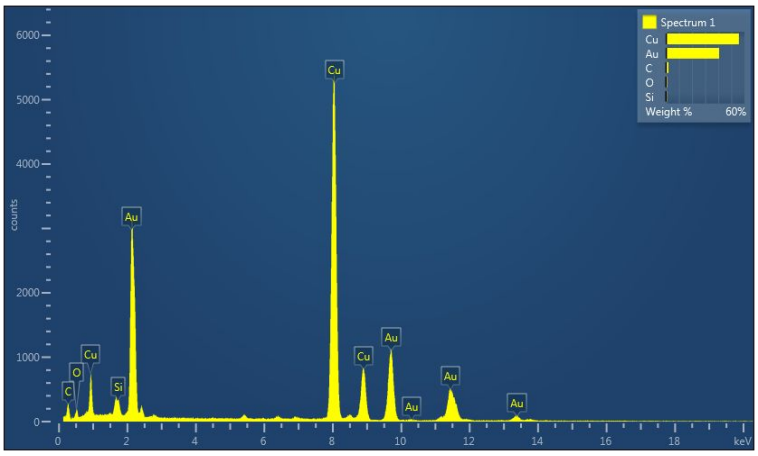

Figure S2. The energy dispersive spectrum of AuNS.

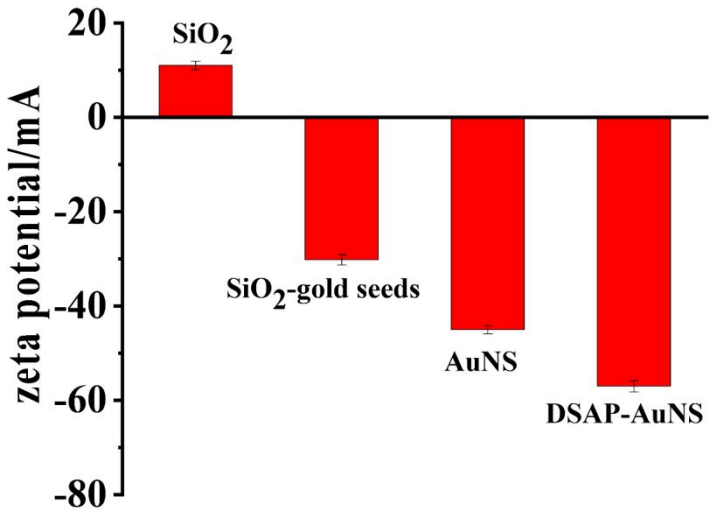

Figure S3. Zeta potential of SiO_2-NH_2, SiO_2-gold seeds, AuNS and DSAP-AuNS.

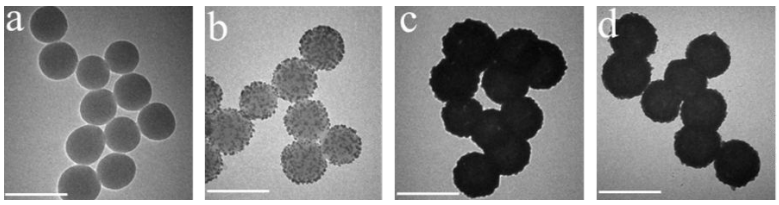

Figure S4. TEM images of SiO_2-NH_2 (a), SiO_2-gold seeds (b), AuNS (c) and DSAP-AuNS (d), scale bar: 200 nm.

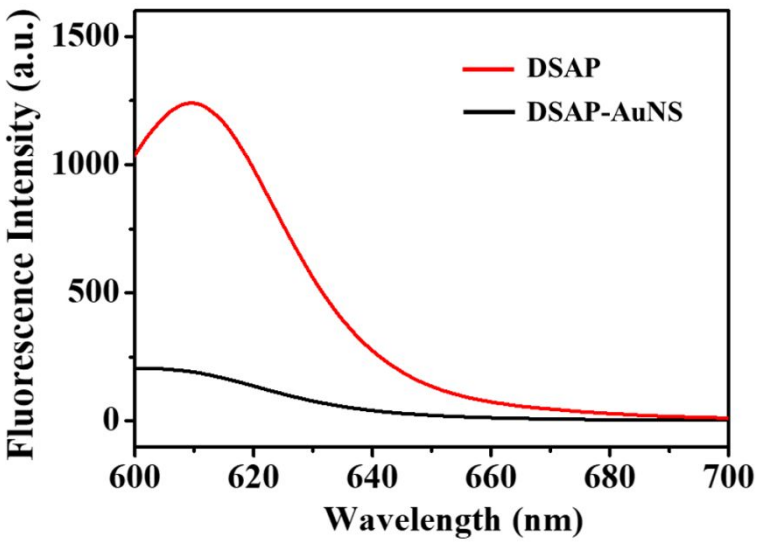

Figure S5. Fluorescence spectra of DSAP and DSAP-AuNS, λex = 590 nm.

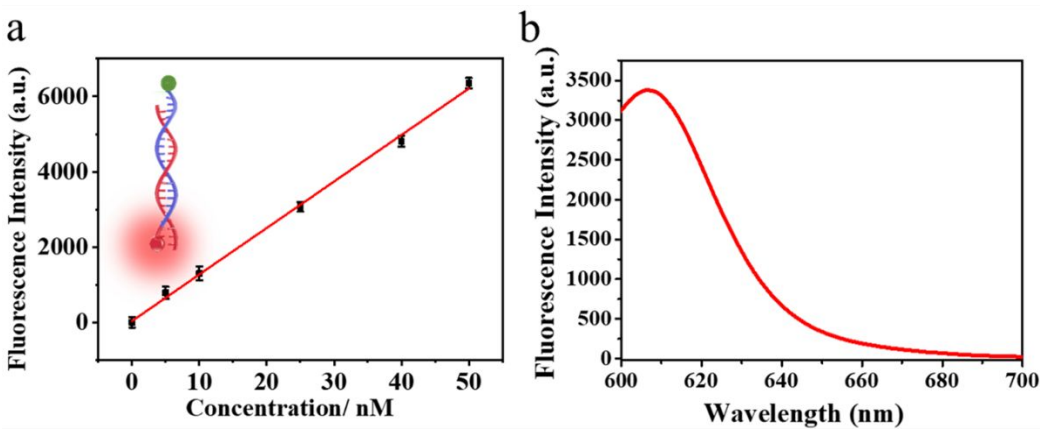

Figure S6. Fluorescence-based method to calculate the number of DSAP loading on AuNS: (a) calibration curve of the fluorescence intensity of Texas-Red versus the concentration of DSAP; (b) Fluorescence emission spectrum of the released DSAP from the ME treated DSAP-AuNS, λex = 590 nm, λem = 612 nm.

As the concentration of DSAP increased, the fluorescence intensity was enhanced in a linear way from 5.00×10^{-9} M to 5.00×10^{-8} M, $I_F = 271.00 + 61.52 \times C_{DSAP}$ (10^{-9} M). Secondly, mercaptoethanol (ME) was brought to the AuNS solution to achieve a final concentration of 20 mM. After incubated overnight with shaking at room temperature, the oligonucleotides were released. Then the released DSAP were separated *via* centrifugation, and the mean fluorescence intensity was measured to be ~3375. So based on the calibration curve, the concentration of released DSAP was calculated to be 2.70×10^{-8} M. And the concentration of AuNS used in our work was 6.0 pM. Therefore, the number of DSAP loaded on each AuNS was calculated to be ~ 4500.

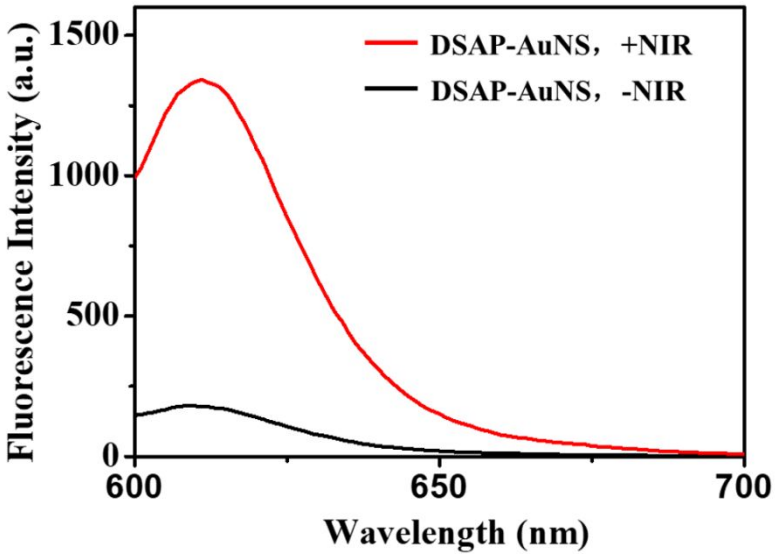

Figure S7. Fluorescence spectra of DSAP-AuNS with (red line) or without NIR irradiation (black line), λex = 590 nm.

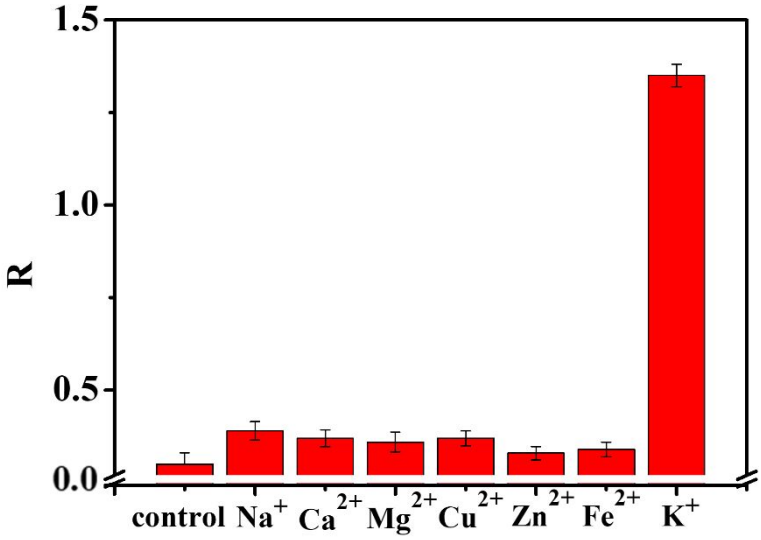

Figure S8. FRET signal R (F_A/F_D) of DSAP-AuNS in the presence of various biological cations at their physiological concentrations: blank, Na^+ (15 mM), Ca^{2+} (2.0 mM), Mg^{2+} (2.0 mM), Cu^{2+} (50 μM), Zn^{2+} (2.0 mM), Fe^{2+} (50 μM), K^+ (150 mM).

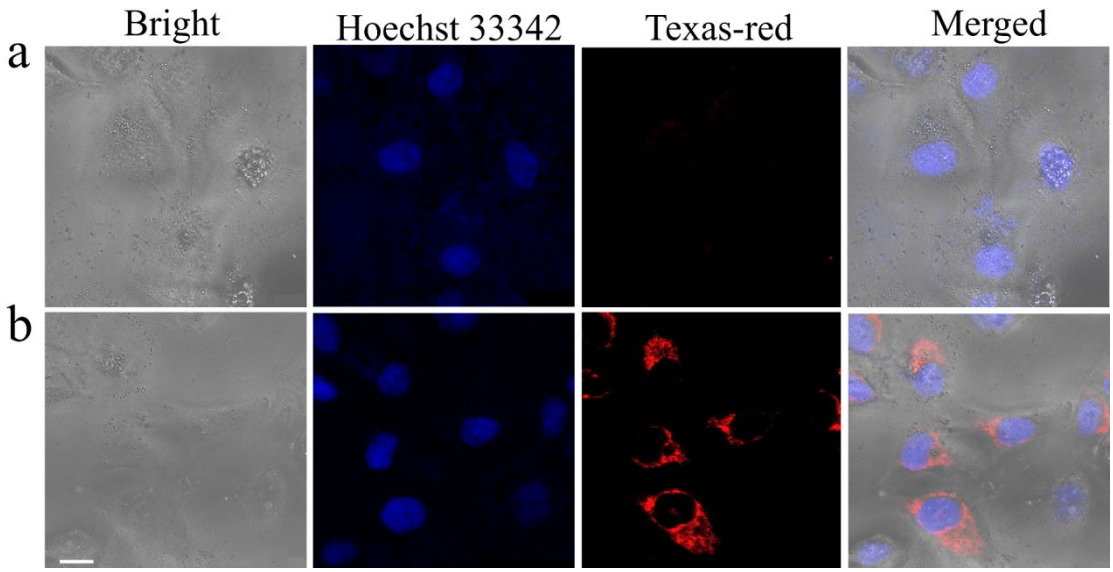

Figure S9. Confocal microscopy images of HeLa cells treated with DSAP (a) or DSAP-AuNS (b) under NIR irradiation, channel of Hoechst 33342, λex: 405 nm, λem: 415-460 nm; channel of Texas Red, λex: 590 nm, λem: 600-640 nm. Scale bar: 10 μm.

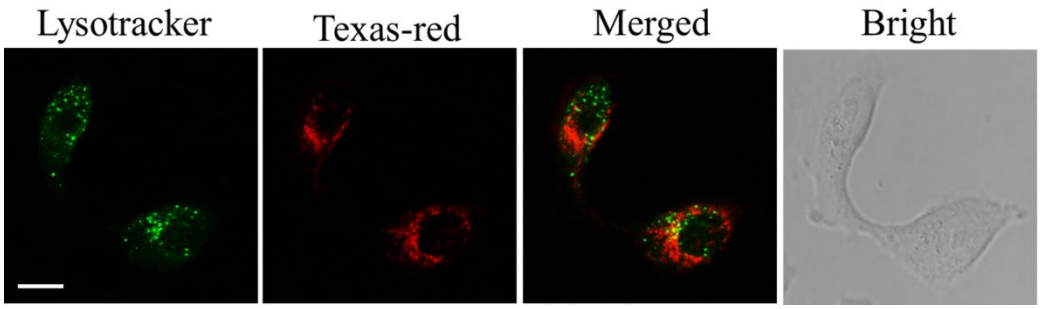

Figure S10. Confocal microscopy images of HeLa cells pretreated with DSAP-AuNS and further stained with Lyso-Tracker GreenDND-26, the red fluorescence from Texas-red shows the location of DSAP-AuNS. The green fluorescence from Lysotracker shows the location of acidic organelles such as lysosome and endosome. Scale bar: 10 μm.

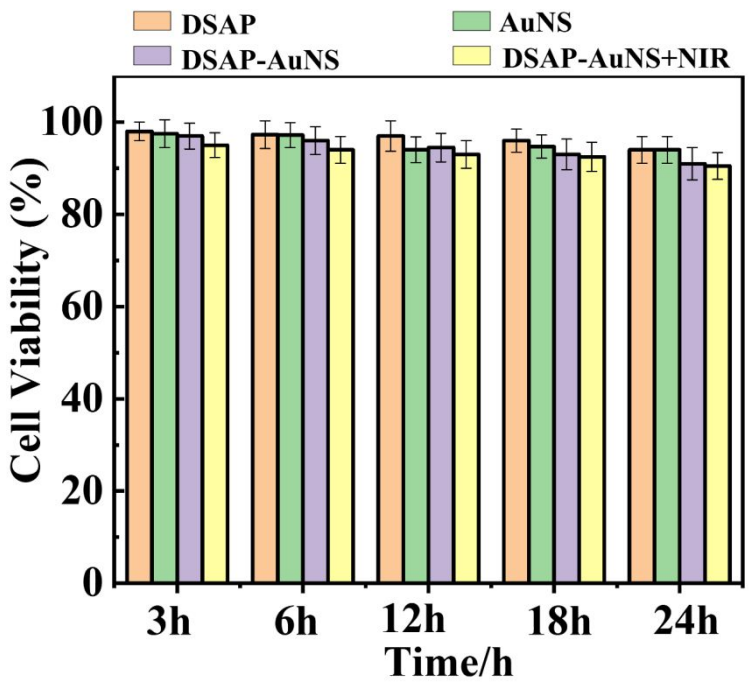

Figure S11. Cell viability of HeLa cells incubated with 14 nM DSAP, 3 pM bare AuNS, 3 pM DSAP-AuNS, 3 pM DSAP-AuNS + NIR irradiation (from the left to right). Error bars represent the standard deviation of three replicates.

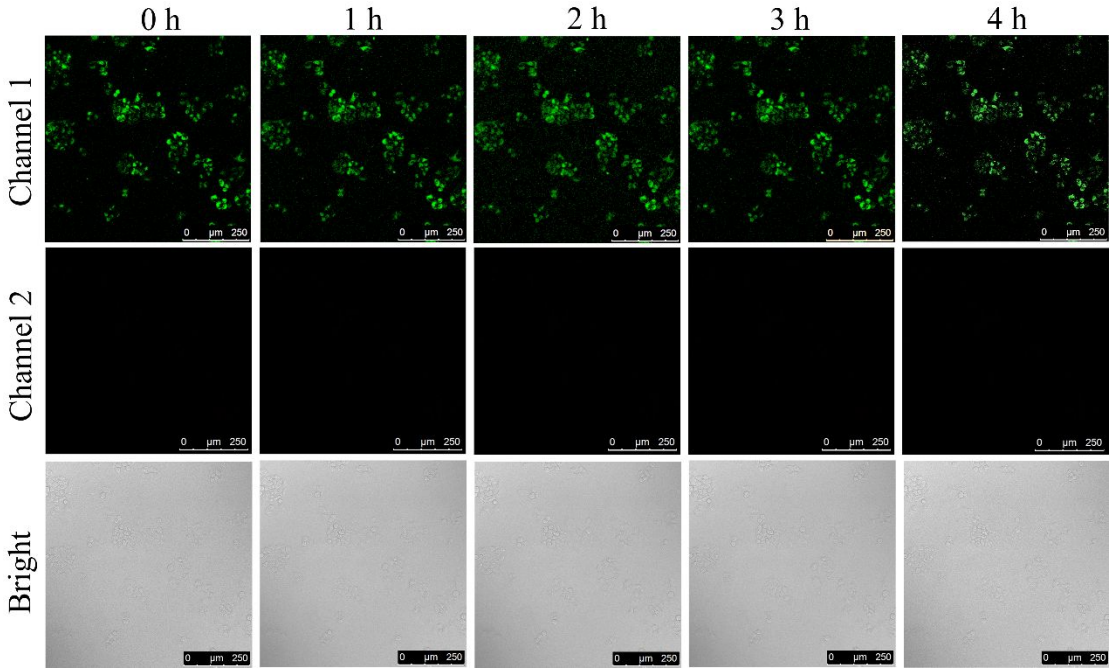

Figure S12. Confocal microscopy images of HeLa cells stand for different times after treated with DSAP-AuNS. Channel 1(Cy3), λ_{ex}: 543 nm, λ_{em}: 555-590 nm; Channel 2 (Texas Red), λ_{ex}: 590 nm, λ_{em}: 600-640 nm. Scale bar: 250 μm.

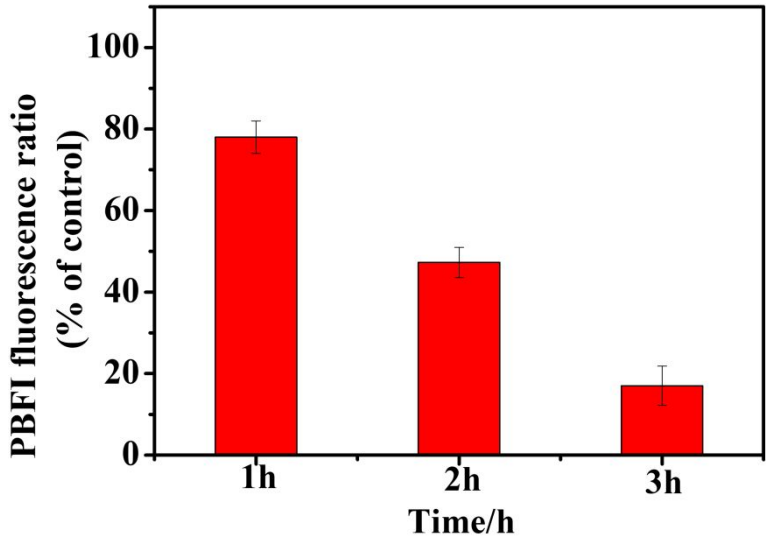

Figure S13. The standard potassium ion assay of HeLa cells treated with reagents (K^+ efflux stimulation amphotericin B, K^+ influx inhibitors bumetanide and ouabain) for different times, the PBFI fluorescence ratio was positively correlated with the intracellular content of K^+.[1]

Reference

(1) Andersson, B.; Janson, V.; Behnam-Motlagh, P.; Henriksson, R.; Grankvist, K. *Toxicol. in Vitro* **2006**, *20,* 986-994.