Supporting Information:

Quantum Dot-Based Sensitization System for Boosted Photon Absorption and Enhanced Second Near-Infrared Luminescence of Lanthanide-Doped Nanoparticle

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Figure S1. Size distribution of the synthesized Er³⁺-doped LNPs.



Figure S2. Size distribution of the synthesized Ag_2S QDs.



Figure S3. XPS spectrum of the synthesized Ag₂S QDs. a) The survey spectra of Ag₂S QDs. b) The corresponding high-resolution spectra of Ag (3d), representing the binding energy of Ag⁺ in Ag₂S. c) The corresponding high-resolution spectra of S (2p), representing the binding energy of S²⁺ in Ag₂S.



Figure S4. XRD pattern of the synthesized Ag_2S QDs. The result shows the position and relative intensity of diffraction peaks of the synthesized Ag_2S QDs were in accordance with those of standard monoclinic Ag_2S (JCPDS card no. 14-0072).



Figure S5. Elemental mapping of Ag₂S QD-sensitized Er^{3+} -doped LNP hybrid including Y, Yb, Er, and Ag elements. The QD-sensitized LNP hybrid was obtained by incubating 60 µL of Er^{3+} -doped LNP (20 mg mL⁻¹) with 600 µL of Ag₂S QDs (OD₈₀₈ = 0.6). Scale bar, 50 nm.



Figure S6. ${}^{2}F_{5/2} \rightarrow {}^{2}F_{7/2}$ (Yb³⁺) emission spectra of QD-sensitized LNP and bare LNP. Emission spectra were obtained from LNP (60 µL, 20 mg mL⁻¹) with/without Ag₂S QDs (25 µL, OD₈₀₈ = 0.6) sensitization at 808 nm (2 W, c.w.) laser excitation.



Figure S7. Fluorescence spectra of QD-sensitized LNP and bare LNP under different laser excitation. Fluorescence spectra were obtained from QD-sensitized LNP at 808 nm (2 W, c.w.) laser excitation, and bare LNP at 980 nm (2 W, c.w.) laser excitation.

Synthesis of ~ 32 nm NaY_{0.795}F₄:Yb_{0.2},Tm_{0.005} core NPs. The procedure is identical to the synthesis of NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core NPs, except using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.795 mmol), Yb(CH₃CO₂)₃ (0.2 mmol), Tm(CH₃CO₂)₃ (0.005 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.78 mmol), Yb(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.02 mmol).



Figure S8. Characterization of the synthesized Tm³⁺-doped LNPs. a) TEM image of the synthesized Tm³⁺-doped LNPs. Scale bar, 50 nm. b) Size distribution of the synthesized Tm³⁺-doped LNPs.



Figure S9. TEM image of Ag₂S QD-sensitized Tm³⁺-doped LNPs system. The relatively largesized Tm³⁺-doped LNPs served as a nanoscale scaffold for decorating multiple copies of Ag₂S QDs. Scale bar, 50 nm.



Figure S10. Fluorescence and absorption spectra of Ag_2S QDs and Tm^{3+} -doped LNPs. Spectra from left to right, coloured according to their transitions in **Figure S12**: Ag_2S QDs adsorption (dashed red line), Ag_2S QDs fluorescence (solid red line) at 808 nm (2 W, c.w.) laser excitation, Tm^{3+} -doped LNPs absorption (dashed blue line) and Tm^{3+} -doped LNPs fluorescence (solid blue line) at 980 nm (2 W, c.w.) laser excitation.



Figure S11. Schematic diagram of proposed energy transfer landscape. The picture shows Ag_2S QDs transfer energy to lanthanide ion pair (Yb^{3+}/Tm^{3+}) in LNP, and emission from LNP.



Figure S12. Fluorescence spectra of QD-sensitized Tm^{3+} -doped LNP system. With increasing amounts of Ag₂S QDs (OD₈₀₈ = 0.6), the emission intensity of Tm^{3+} -doped LNPs (60 µL, 20 mg mL⁻¹) increased under 808 nm (2 W, c.w.) laser excitation.

Synthesis of Ag₂Se QDs. 0.1 mmol of CH₃COOAg, 286 μ L of 1-octanethiol was added into 5 mL of ODE in a 50 mL three-neck bottom flask under argon atmosphere. Subsequently, 0.1 mmol of Se dissolved in TOP (1.5 mL) was injected rapidly into the mixture at 160 °C under vigorous stirring. The reaction then was quickly terminated in an ice-water bath. The resulting product was precipitated by addition of acetone, collected by centrifugation at 10000 rpm for 3 min, and then dispersed in 1 mL of cyclohexane. Finally, the obtained Ag₂Se QDs was mixed with an ethanol solution containing mercaptoundecanoic acid. After stirred overnight at room temperature, the resulting product was centrifuged at 10000 rpm for 20 min, and washed with ethanol/deionized water several times. The product was redispersed in Tris-HCl buffer (0.01 M, pH 7.4), and stored at 4 °C in the dark for further use.



Figure S13. Characterization of the synthesized $Ag_2Se QDs. a$) TEM image of the synthesized $Ag_2Se QDs.$ Scale bar, 25 nm. b) Size distribution of the synthesized $Ag_2Se QDs.$



Figure S14. Fluorescence and absorption spectra of Ag_2Se QDs and Er^{3+} -doped LNPs. Spectra from left to right: Ag_2Se QDs adsorption (dashed red line), Ag_2Se QDs fluorescence (solid red line) at 808 nm (2 W, c.w.) laser excitation, Er^{3+} -doped LNPs absorption (dashed blue line) and Er^{3+} -doped LNPs fluorescence (solid blue line) at 980 nm (2 W, c.w.) laser excitation.



Figure S15. Fluorescence spectra of Ag₂Se QD-sensitized Er^{3+} -doped LNP system. With increasing amounts of Ag₂Se QDs (OD₈₀₈ = 0.6), the emission intensity of Er^{3+} -doped LNPs (60 µL, 20 mg mL⁻¹) increased under 808 nm (2 W, c.w.) laser excitation.

Synthesis of ~ 32 nm NaY_{0.785}F₄:Nd_{0.01},Yb_{0.2},Er_{0.005} core NPs. The procedure is identical to the synthesis of NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core NPs, except using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.785 mmol), Nd(CH₃CO₂)₃ (0.01 mmol), Yb(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.005 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.78 mmol), Yb(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.2 mmol), Er(CH₃CO₂)₃ (0.2 mmol).



Figure S16. Characterization of the synthesized Nd³⁺-doped LNPs. a) TEM image of the synthesized Nd³⁺-doped LNPs. Scale bar, 50 nm. b) Size distribution of the synthesized Nd³⁺-doped LNPs.



Figure S17. Fluorescence spectra of Ag₂S QD-sensitized Nd³⁺-doped LNP system. With increasing amounts of Ag₂S QDs (OD₈₀₈ = 0.6), the emission intensity of Nd³⁺-doped LNPs (60 μ L, 20 mg mL⁻¹) increased under 808 nm (2 W, c.w.) laser excitation.



Figure S18. Fluorescence images of bare Nd³⁺-doped LNP (600 μ L, 2 mg mL⁻¹) and Yb³⁺-doped LNP (600 μ L, 2 mg mL⁻¹) under 808 nm (2 W, c.w.) laser excitation.

Determination of ions concentrations by ICP-MS. 2 μ L of the QD-sensitized LNP hybrid solution was added into 500 μ L of aqua regia (HCl:HNO₃ = 3:1, v/v), stirring overnight at room temperature. The final volume of solution was filled to 10 mL with DI water for ICP-MS analysis.

Determination the average surface density of Ag₂S QDs on the surface of LNP. First, we need to estimate the number of LNPs in QD-sensitized LNP hybrid solution. For this purpose, we assume a single LNP to be spherical with volume (V_{LNP}) described by:

$$V_{LNP} = \frac{4}{3}\pi r_{LNP}^{3},$$
 (1)

where r_{LNP} is the radius of the LNP. Since the LNP consists of hexagonal unit cells (Fig. S18), volume of a hexagonal unit cell (uV_h) is described by:

$$uV_h = \frac{2\sqrt{3}}{4}a_h^2 c_h,\tag{2}$$

where a_h and c_h are lattice parameters of hexagonal unit cells (hexagonal lattice parameters $a_h = 0.596$ nm, $c_h = 0.353$ nm). Thus, the number of unit cells in an LNP can be given by:

$$uN_h = V_{LNP}/uV_h. \tag{3}$$

This calculation are based on the assumption that the effects of crystal dislocations and rounding error in the total number of unit cells to be negligible, and that lattice parameters are accurate, as well as LNPs are composed of 100% hexagonal unit cells.

If no lanthanide ions were doped in LNPs, the Yttrium (Y) atomic weight of a single hexagonal $Na_{1.5}Y_{1.5}F_6$ unit cell (uAW_Y) is described by:

$$uAW_Y = 1.5 \times AW_Y,\tag{4}$$

where AW_Y is the atomic weight (g mol⁻¹) of Y. We assume any mass difference due to loss

of electrons in ionisation to be negligible. Considering a fraction of Y^{3+} ions are substituted by lanthanide ions during Er^{3+} -doped LNPs synthesis, we intrudce a factor ($f_{Y,LNP}$) as the fractional percentage of Y^{3+} ions in Er^{3+} -doped LNPs unit cells to describe the Y atomic weight of a single unit cell within Er^{3+} -doped LNPs:

$$uAW_{Y,LNP} = 1.5 \times f_{Y,LNP} \times AW_Y. \tag{5}$$

According to eq 3 and 5, the average weight of Y^{3+} ions in a single LNP ($W_{Y,LNP}$) can be described by

$$W_{Y,LNP} = 1.5 \times f_{Y,LNP} \times AW_Y \times uN_h. \tag{6}$$

According to eq 6, the number of LNPs in QD-sensitized LNP system (N_{LNP}) can be estimated as

$$N_{LNP} = W_{Y,system}/W_{Y,LNP},\tag{7}$$

where $W_{Y,system}$ is the weight of Y³⁺ ions in the total system that obtained by ICP-MS (Fig. S14).

Subsequently, we also estimate the number of Ag₂S QDs in QD-sensitized LNP system solution by the similar method. In this case, we assume Ag₂S QDs is spherical with a radius of r_{QD} , and the volume of a single QD (V_{QD}) can be described by:

$$V_{QD} = \frac{4}{3}\pi r_{QD}{}^3,$$
 (8)

Considering the synthesized Ag₂S QDs belong to monoclinic crystal (Fig. S4), volume of a monoclinic unit cell (uV_m) is given by:

$$uV_m = a_m b_m c_m \sin\beta, \tag{9}$$

where a_m , b_m and c_m are lattice parameters of monoclinic unit cells (monoclinic lattice parameters $a_m = 0.423$ nm, $b_m = 0.693$ nm, $c_m = 0.786$, and $\beta = 99.61^\circ$). Thus, the number of unit cells in a QD can be estimated by:

$$uN_m = V_{QD}/uV_m. \tag{10}$$

This calculation are also based on the assumption that the effects of crystal dislocations and rounding error in the total number of unit cells to be negligible, and that lattice parameters are accurate.

Accodring to the lattice parameters of Ag₂S QDs, the silver (Ag) atomic weight of a single monoclinic Ag₈S₄ unit cell (uAW_{Ag}) can be described by:

$$uAW_{Ag} = 8 \times AW_{Ag},\tag{11}$$

where AW_{Ag} is the atomic weight (g mol⁻¹) of Ag.

According to eq 10 and 11, the average weight of Ag^+ ions in a single QD ($W_{Ag,QD}$) can be described by

$$W_{Aq,OD} = 8 \times AW_{Aq} \times uN_m. \tag{12}$$

According to eq 12, the number of QDs in QD-sensitized LNP system (N_{QD}) can be calculated as

$$N_{QD} = W_{Ag,system}/W_{Ag,QD},$$
(13)

where $W_{Ag,system}$ is the weight of Ag⁺ ions in the total system that obtained by ICP-MS (Fig. S17).

Finally, through combining the eq 7 and 13, the average surface density of Ag_2S QDs on the surface of a single LNP can be estimated by

$$Density = \frac{N_{QD}}{N_{LNP}}$$
(14)

Calculation of ion contents in the shell of LNPs. The molar content of two rare earth ions

 $(Y^{3+} and Yb^{3+})$ in the shell region of core-shell LNPs can be calculated by

$$n_{s,i} = n_{cs,i} - n_{c,i} \tag{15}$$

where $n_{cs,i}$ is the total molar content of rare earth ions ($i = Y^{3+}$ and Yb^{3+}) in core-shell LNPs, $n_{c,i}$ and $n_{s,i}$ are the molar content of rare earth ions ($i = Y^{3+}$ and Yb^{3+}) in the core and shell region of core-shell LNPs, respectively. The molar content of rare earth ions was determined by ICP-MS (Fig. S14-16).

According to the eq 15, the molar ratio of two rare earth ions $(Y^{3+} \text{ and } Yb^{3+})$ in the shell region of core-shell LNPs can be calculated by

$$Y\% = \frac{n_{s,Y}}{n_{s,Y} + n_{s,Yb}}$$
(16)

and

$$Yb\% = \frac{n_{s,Yb}}{n_{s,Y} + n_{s,Yb}}$$
(17)



Figure S19. Working standard curve for quantification of the concentration of Y^{3+} ions. The Linearity of working standard curve for Y^{3+} ions was determined in the range of 0.1-10 µg mL⁻¹.



Figure S20. Working standard curve for quantification of the concentration of Yb^{3+} ions. The Linearity of working standard curve for Yb^{3+} ions was determined in the range of 0.1-10 µg mL⁻¹.



Figure S21. Working standard curve for quantification of the concentration of Er^{3+} ions. The Linearity of working standard curve for Er^{3+} ions was determined in the range of 0.1-10 µg mL⁻¹.



Figure S22. Working standard curve for quantification of the concentration of Ag^+ ions. The Linearity of working standard curve for Ag^+ ions was determined in the range of 0.1-10 µg mL⁻¹.



Figure S23. XRD patterns of core and core-shell LNPs. The result shows the position and relative intensity of diffraction peaks of all the synthesized core and core-shell LNPs were in accordance with those of standard NaYF₄ (JCPDS card no. 16-0334).

Synthesis of ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~1.9 nm NaYF₄ shell. An aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol) was added into a 50 mL three-necked flask containing OA (3 ml) and ODE (7 ml). The mixture was heated to 160 °C for 1 h, and then naturally cooled down to room temperature. After that 2 mL of the prepared ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core (~0.2 mmol) in cyclohexane was added along with 6 mL methanol solution containing 0.2 mmol NH₄F and 0.125 mmol NaOH, the mixture was stirred for 30 min at 50 °C. Subsequently, the temperature was increased to 110 °C to completely remove methanol from the mixture, and the resulting mixture was further heated to 300 °C for 1.5 h under an argon atmosphere. Then, the reaction mixture was naturally cooled down to room temperature. The obtained nanoparticles were precipitated by addition of ethanol, collected by centrifugation at 6000 rpm for 10 min, then washed with cyclohexane and ethanol several times, and finally dispersed in 2 mL of cyclohexane.

Synthesis of ~32nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~3.2 nm, ~5.1 nm and ~6.7 nm NaYF₄ shell. The procedure is identical to the synthesis of \sim 32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~1.9 nm NaYF₄ shell, except using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.1 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), and methanol solution (6 mL) containing 0.4 mmol NH₄F and 0.25 mmol NaOH to replace methanol (6 mL) solution containing 0.2 mmol NH₄F and 0.125 mmol NaOH, respectively, for ~32 nm $NaY_{0.8}F_4$: $Yb_{0.18}$, $Er_{0.02}$ core with ~3.2 nm $NaY_{0.9}F_4$: $Yb_{0.1}$ shell; using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.15 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), and methanol solution (6 mL) containing 0.6 mmol NH₄F and 0.375 mmol NaOH to replace methanol (6 mL) solution containing 0.2 mmol NH₄F and 0.125 mmol NaOH, respectively, for ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~5.1 nm NaY_{0.8}F₄:Yb_{0.2} shell; and using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.2 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), and methanol solution (6 mL) containing 0.8 mmol NH₄F and 0.5 mmol NaOH to replace methanol (6 mL) solution containing 0.2 mmol NH₄F and 0.125 mmol NaOH, respectively, for ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~6.7 nm NaY_{0.8}F₄:Yb_{0.2} shell.



Figure S24. Characterization of the synthesized core-shell LNPs. a) TEM image of LNPs with ~1.9 nm shell. b) Size distribution of LNPs with ~1.9 nm shell. c) TEM image of LNPs with ~3.2 nm shell. d) Size distribution of LNPs with ~3.2 nm shell. e) TEM image of LNPs with ~5.1 nm shell. f) Size distribution of LNPs with ~5.1 nm shell. g) TEM image of LNPs with ~6.7 nm shell. h) Size distribution of LNPs with ~6.7 nm shell. Scale bar, 50 nm.

Synthesis of ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~1.7 nm NaY_{1-x}F₄:Yb_x (x = 0.1, 0.2) shell. The procedure is identical to the synthesis of ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~1.9 nm NaYF₄ shell, except using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.045 mmol), Yb(CH₃CO₂)₃ (0.005 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol) for ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02} core with ~1.7 nm NaY_{0.9}F₄:Yb_{0.1} shell, and using an aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.04 mmol), Yb(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.04 mmol), Yb(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), Yb(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (1.2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), Yb(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (1.2 mL) of Y(CH₃CO₂)₃ (0.05 mmol), Yb(CH₃CO₂)₃ (0.01 mmol) to replace the aqueous solution (0.2 mL) of Y(CH₃CO₂)₃ (0.05 mmol) for ~32 nm NaY_{0.8}F₄:Yb_{0.18},Er_{0.02}



Figure S25. Characterization of the synthesized core LNPs with Yb^{3+} doping shell. a) TEM image of LNPs with NaYF₄ shell. b) Size distribution of LNPs with NaYF₄ shell. c) TEM image of LNPs with 10% Yb^{3+} doping NaYF₄ shell. d) Size distribution of LNPs with 10% Yb^{3+} doping NaYF₄ shell. e) TEM image of LNPs with 20% Yb^{3+} doping NaYF₄ shell. f) Size distribution of LNPs with 20% Yb^{3+} doping NaYF₄ shell. f) Size distribution of LNPs with 20% Yb^{3+} doping NaYF₄ shell. f) Size distribution of LNPs with 20% Yb^{3+} doping NaYF₄ shell. Scale bar, 50 nm.



Figure S26. Fluorescence and absorption spectra of IR-806 dye and Er³⁺-doped LNPs. Spectra from left to right: IR-806 adsorption (dashed red line), IR-806 fluorescence (solid red line) at 808 nm (2 W, c.w.) laser excitation, Er³⁺-doped LNPs absorption (dashed blue line) and Er³⁺-doped LNPs fluorescence (solid blue line) at 980 nm (2 W, c.w.) laser excitation.



Figure S27. Fluorescence spectra of dye-sensitized Er^{3+} -doped LNP system. With increasing amounts of IR-806 dye (0.1 µg mL⁻¹), the emission intensity of Er^{3+} -doped LNPs (60 µL, 20 mg mL⁻¹) increased under 808 nm (2 W, c.w.) laser excitation.



Figure S28. Illustration of preparation scheme for Ag₂S QDs@mal.



Figure S29. FI-TR spectrum of the synthesized Ag₂S QDs@mal. The typical bands at 2922 cm⁻¹ and 2852 cm⁻¹ are assigned to the asymmetric and symmetric stretching vibrations of C-H of MPA. The sharp and strong band at 1716 cm⁻¹ is assigned to the C=O stretching vibrations of carboxylic acid group. After react with NH₂-PEG₂-maleimide, the new bands at 1631 cm⁻¹ (C=C vibrations), 1774 cm⁻¹ and 1701 cm⁻¹ (C=O vibrations) correspond to the characteristic bands of maleimide group. The results confirm the successful preparation of Ag₂S QDs@mal.



Figure S30. Illustration of preparation scheme for LNPs@RP.



Figure S31. FI-TR spectrum of the synthesized LNPs@RP. The results show the characteristic bands of oleic acid at 3005 cm⁻¹ (C-H stretch in C=C-H), 2924 cm⁻¹ and 2854 cm⁻¹ (CH₂ asymmetric and symmetric stretch), 1560 cm⁻¹ and 1466 cm⁻¹ (COO⁻ asymmetric and symmetric stretch) disappear after HCl treatment. After react with COOH-PEG₂-maleimide, the new bands at 1570 cm⁻¹ (C=C vibrations), 1775 cm⁻¹ and 1701 cm⁻¹ (C=O vibrations) correspond to the characteristic bands of maleimide group. When further covalently attach a peptide sequence, the relative intensity of band at 1570 cm⁻¹ decreased, which confirms the successful preparation of LNPs@RP.



Figure S32. *Ex vivo* fluorescence spectra based on QD-sensitized LNP system upon GSH treatment. (a) Fluorescence spectra of LNPs@RP (60 μ L, 20 mg mL⁻¹) with different amounts of Ag₂S QDs@mal (0-1000 μ L, OD₈₀₈ = 0.6) upon the addition of GSH solution (10 μ L, 1.0 M) at 808 nm (2 W, c.w.) laser excitation. (b) Histogram analysis of fluorescence intensity changes in (a).

For investigating the tumor retention kinetic of $Ag_2S@mal QDs$, 400 µL of $Ag_2S@mal QDs$ ($OD_{808} = 3.0$) was intravenously injected into the tumor-bearing mouse, and *in vivo* imaging was performed under a 808 nm diode laser excitation light with a band pass filter (1000-1100 nm).



Figure S33. Tumor retention kinetics of $Ag_2S@mal QDs$. The result shows the maximum signal intensity of $Ag_2S QDs$ in the tumor area is ~10 min of post-injection.

For investigating the tumor retention kinetic of LNPs@RP, 200 µL of LNPs@RP (20 mg mL⁻¹) was intravenously injected into the tumor-bearing mouse, and *in vivo* imaging was performed under a 980 nm diode laser excitation light with a band pass filter (1500-1600 nm).

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Figure S34. Tumor retention kinetics of LNPs@RP. The result shows the maximum signal intensity of LNPs@RP in the tumor area is ~36 h of post-injection.

Synthesis of PbS QDs. 1 mL of $Pb(CH_3CO_2)_2$ aqueous solution (0.1 M) and 2 mL of GSH aqueous solution (0.1 M) were added into 25 mL of deionized water in a 50 mL round bottom flask under argon atmosphere. The pH of the reaction solution was adjusted with NaOH aqueous solution (1.0 M). Subsequently, 0.1 mL of Na₂S aqueous solution (0.1 M) was slowly added with a small volume syringe. The color of solution immediately turned to clear brown, indicate the formation of PbS QDs. The obtained PbS QDs was stored at 4 °C in the dark for further use.



Figure S35. Characterization of the synthesized PbS QDs. a) TEM image of the synthesized PbS QDs. Scale bar, 25 nm. b) Size distribution of the synthesized PbS QDs.



Figure S36. Fluorescence and absorption spectra of PbS QDs and Er^{3+} -doped LNPs. Spectra from left to right: PbS QDs adsorption (dashed red line), PbS QDs fluorescence (solid red line) at 808 nm (2 W, c.w.) laser excitation, Er^{3+} -doped LNPs absorption (dashed blue line) and Er^{3+} -doped LNPs fluorescence (solid blue line) at 980 nm (2 W, c.w.) laser excitation.



Figure S37. Fluorescence spectra of PbS QD-sensitized Er^{3+} -doped LNP system. With increasing amounts of PbS QDs (OD₈₀₈ = 0.6), the emission intensity of Er^{3+} -doped LNPs (60 μ L, 20 mg mL⁻¹) increased under 808 nm (2 W, c.w.) laser excitation. Quantum yield of PbS QDs is ~16%.