Supporting Information for

Triple-functional Core-Shell Structured Upconversion Luminescent Nanoparticles Covalently Grafted by Photosensitizer for Luminescent, Magnetic Resonance Imaging and Photodynamic Therapy *in Vitro*

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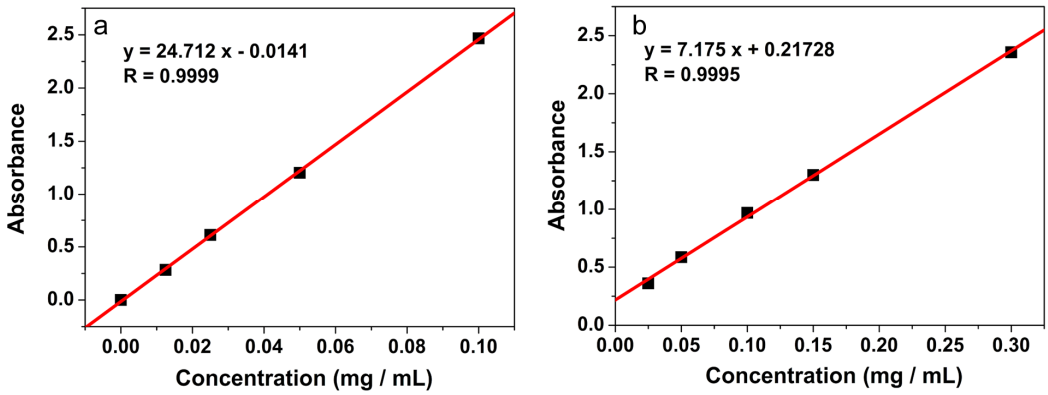

Figure S1. The concentration-dependent ultraviolet-visible absorption signal of PS molecule in DMF solution. ((a) based on the typical absorption of HP at about 400 nm and (b) based on the typical absorption of SPCD at about 668 nm).

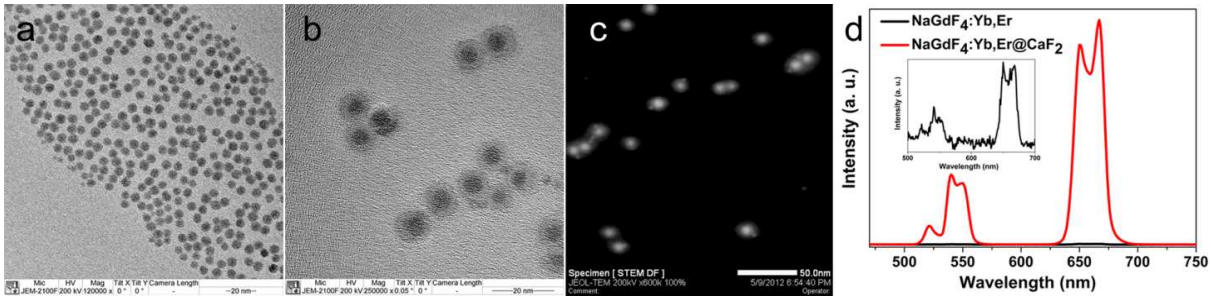

Figure S2. The HRTEM image of $NaGdF_4$:Yb,Er NPs (a) and $NaGdF_4$:Yb,Er@CaF_2 NPs (b), HAADF-STEM image of $NaGdF_4$:Yb,Er@CaF_2 NPs (c) and upconversion luminescent spectra of $NaGdF_4$:Yb,Er and $NaGdF_4$:Yb,E @CaF_2 NPs (d).

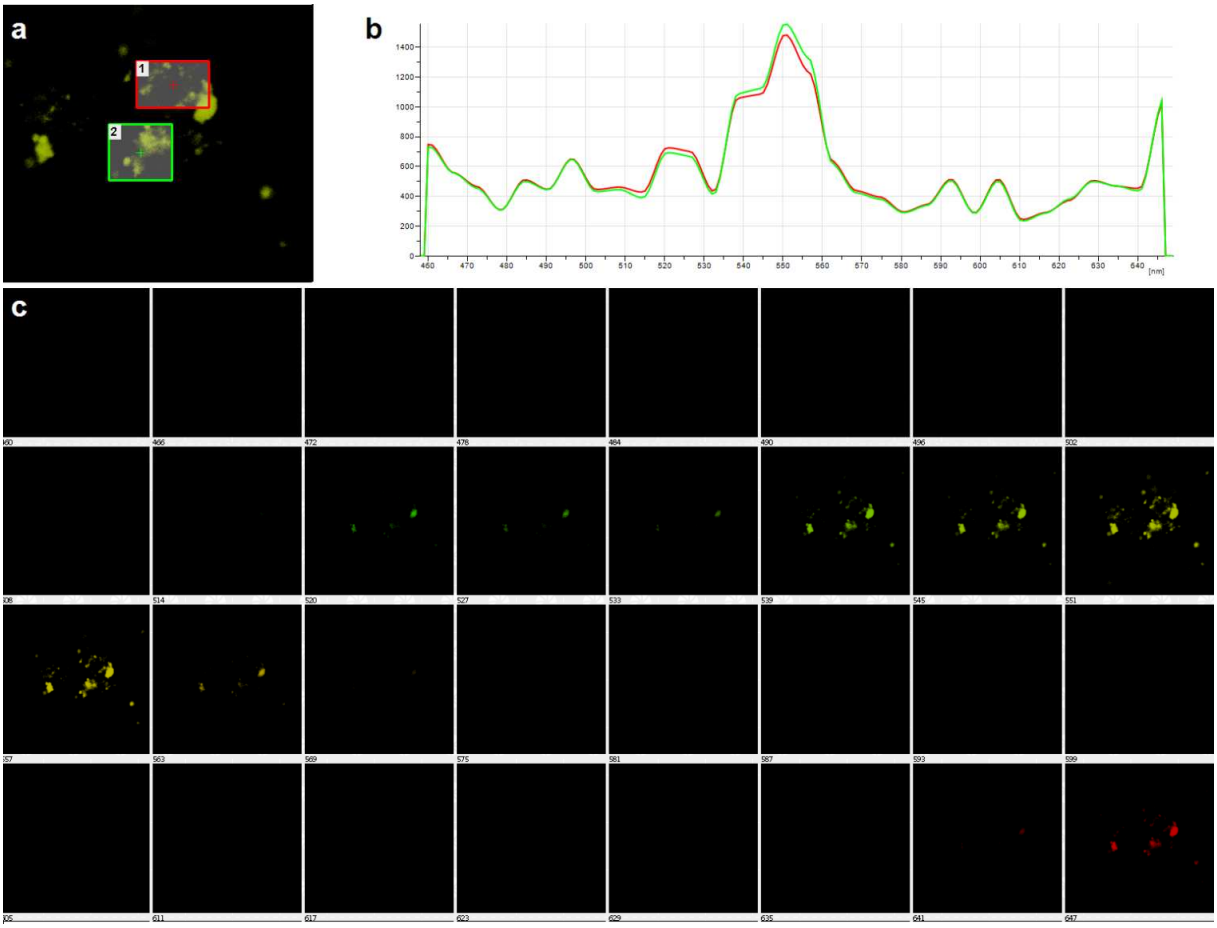

Figure S3. The HeLa cells incubated with as-prepared nanomaterial $NaGdF_4$:Yb,Er@CaF_2-SPCD and the corresponding upconversion emission spectra (b). The red and green lines correspond to the two areas. Fluorescent images acquired from 32 channels in 458-650 nm region (c). The number under each panel corresponds to the detected central spectral wavelength.

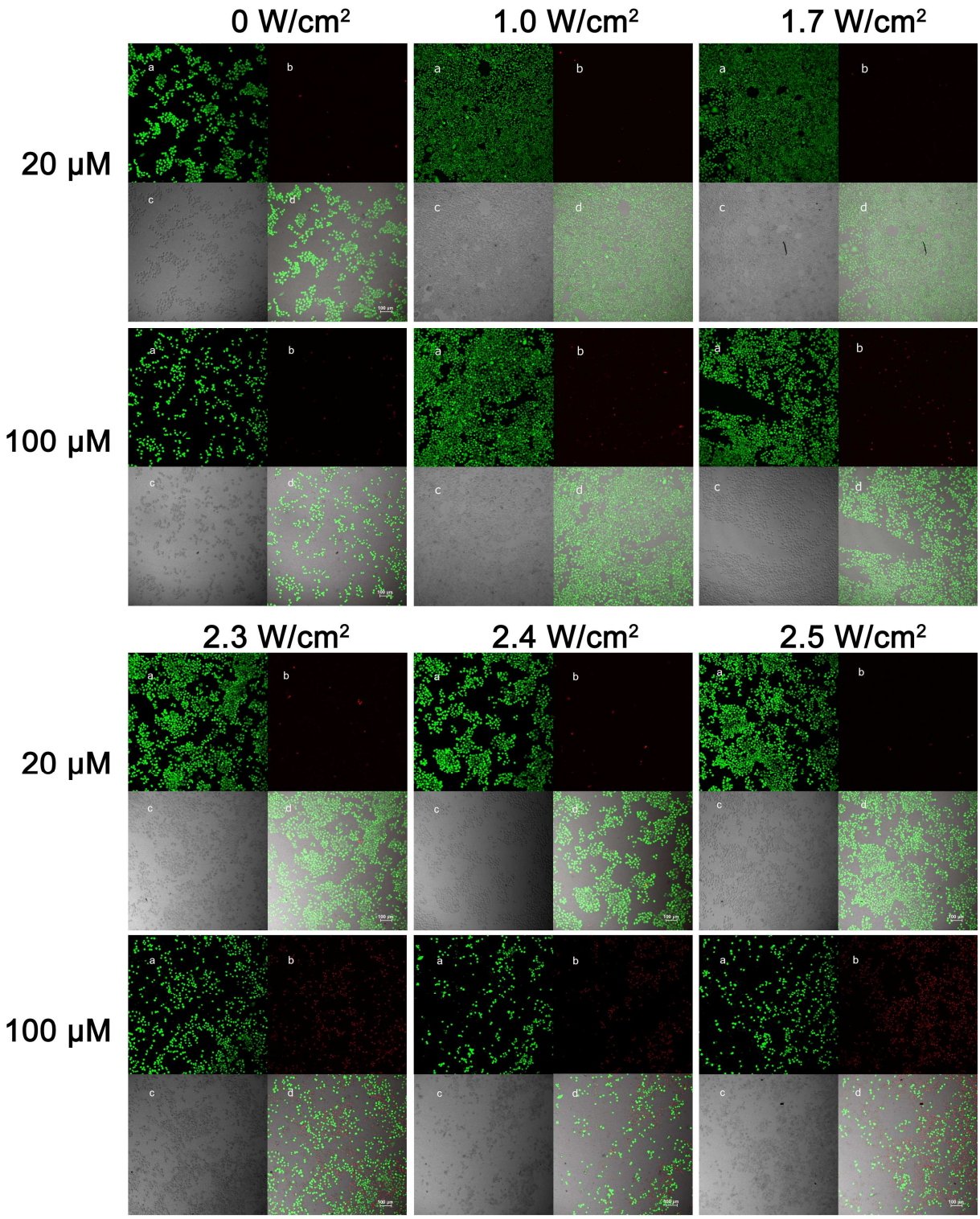

Figure S4. Confocal fluorescent images of HeLa cells incubated with as-prepared nanomaterial $NaGdF_4$:Yb,Er@CaF_2@SiO_2-SPCD of 0 and 100 μM after 5 min irradiation with power density of 0, 1.0, 1.7 2.3, 2.4 and 2.5 W/cm^2 under 980 nm laser diode (in each sample a for fluorescent channel of CAM, b for channel of PI, c for bright-field and d for merged images of HeLa cells).

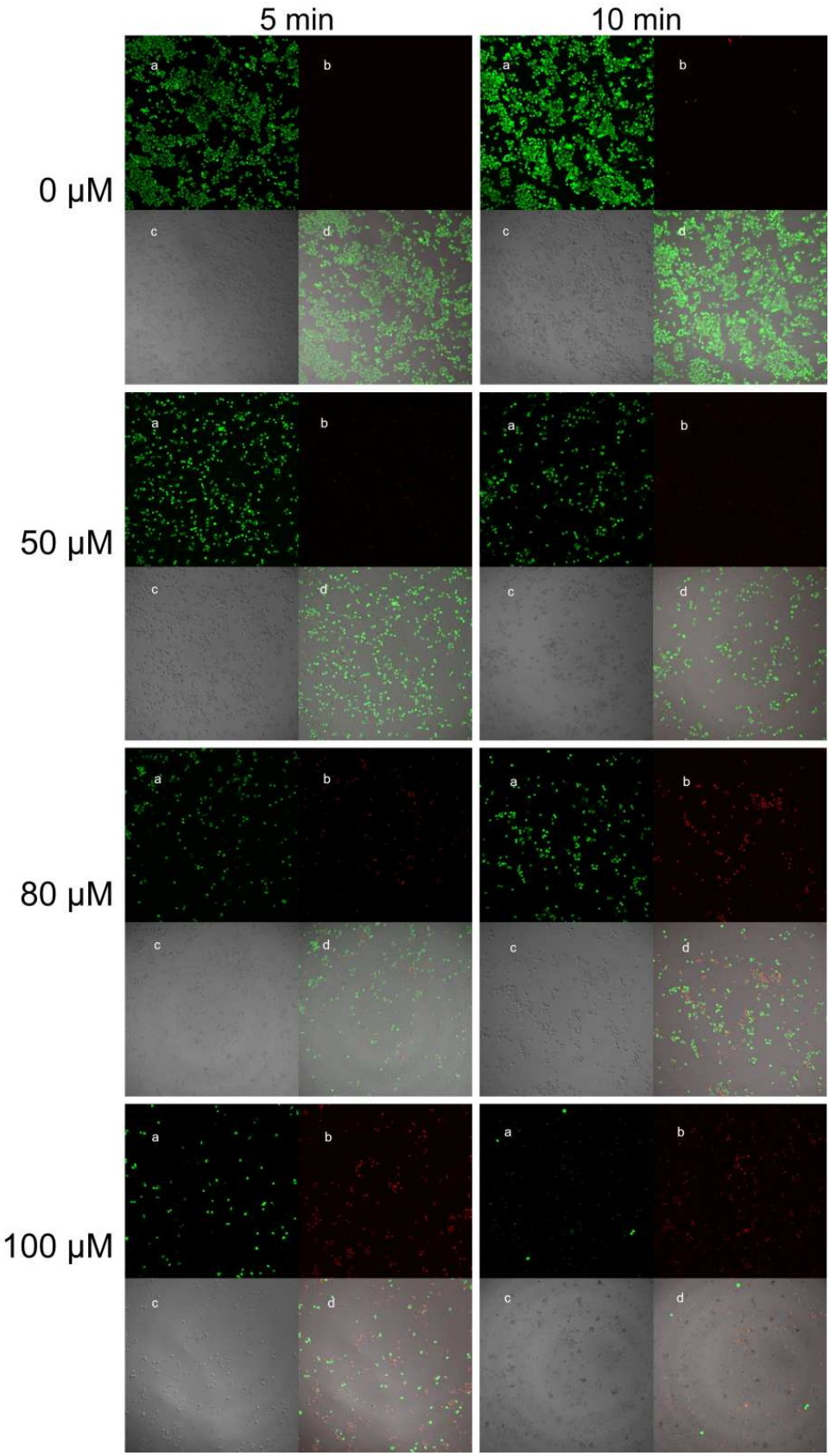

Figure S5. Confocal fluorescent images of HeLa cells incubated with as-prepared nanomaterial $NaGdF_4$:Yb,Er@CaF_2@SiO_2-SPCD with the concentration of 0, 50, 80 and 100 μM after 980 nm laser irradiation within 5 or 10 min with power density of 2.5 W/cm^2 (in each sample a for fluorescent channel of CAM, b for channel of PI, c for bright-field and d for merged images of HeLa cells).

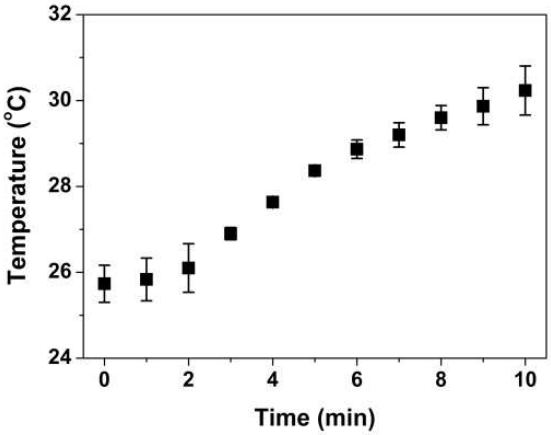

Figure S6. The time-dependent temperature in HeLa cell medium under the irradiation of 980 nm laser diode with power density of 2.5 W/cm^2.

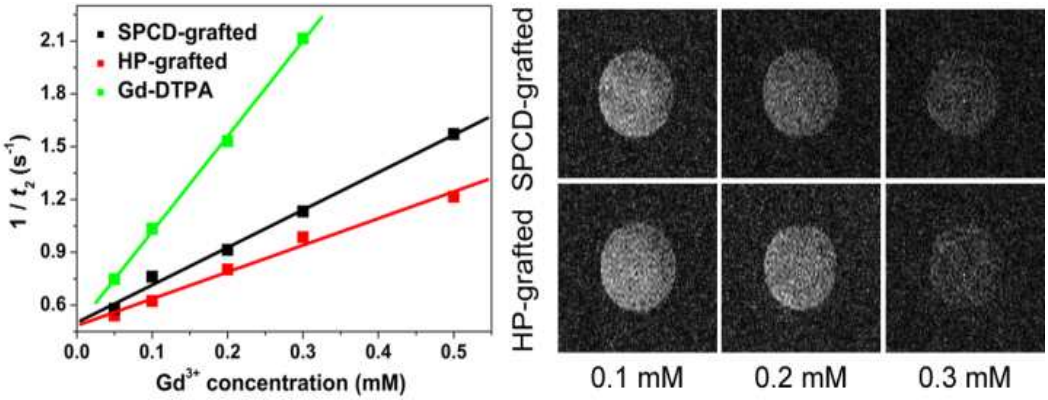

Figure S7. Transverse relaxivities (r_2) data (fitting curve) of as-prepared nanomaterials ($NaGdF_4$:Yb,Er@CaF_2@SiO_2-PS) and commercial contrast agent Gd-DTPA aqueous solution, and t_2-weighted MR images of nanomaterials aqueous solution with Gd^{3+} ion concentration from 0.1 to 0.3 mM (D4= 200 ms).

Table S1. The electronic longitudinal (t_1) and transverse relaxation (t_2) time of as-prepared nanomaterial $NaGdF_4$:Yb,Er@CaF_2@SiO_2-HP, $NaGdF_4$:Yb,Er@CaF_2@SiO_2-SPCD and commercial contrast agent (Gd-DTPA). The last row for the ratio of longitudinal relaxivities value (r_1) to transverse relaxivities value (r_2).

Gd^{3+} (mM)	Gd-DTPA		HP-grafted nanomaterial		SPCD-grafted nanomaterial	
	t_1 (ms)	t_2 (ms)	t_1 (ms)	t_2 (ms)	t_1 (ms)	t_2 (ms)
0.05	/	1339	2401	1847	2103	1728
0.10	1163	968	1931	1605	1395	1314
0.15	/	/	1626	/	1186	/
0.20	755	653	1257	1246	975	1094
0.30	567	473	/	1015	/	883
0.50	/	/	/	823	/	637
r_1 / r_2	0.85		1.62		1.67	