

Figure S1. The size distribution histograms of CuFeSe<sub>2</sub>. The particle size distribution of CuFeSe<sub>2</sub>, counted from 202 nanocrystals shown in typical TEM images, showing these nanoparticles are with small size and their particle sizes were relatively uniform.



Figure S2. Energy dispersive spectrometer of the as-synthesized CuFeSe<sub>2</sub>-PEG-FA nanoparticles, identifying the presence of Cu, Fe and Se elements.



Figure S3. (a) Ultraviolet-visible (UV-Vis)-NIR absorbance (inset: photographs of different solutions) and (b) photograph of an aqueous solution of CuFeSe<sub>2</sub>-PEG-FA NCs ( $50 \mu g/mL$ ) stored for 15 days under ambient conditions.



Figure S4. The linear correlation between the CT values of CuFeSe<sub>2</sub>-PEG-FA nanocrystals with their concentrations respectively.



Figure S5. In vitro relative cell viabilities of 4T1 and HepG2 cells after incubation with various concentrations of CuFeSe<sub>2</sub>-PEG-FA (a) and CuFeSe<sub>2</sub> (b) nanoparticles for 24 h. The relative cell viability (%) values are an average of at least three independent experiments  $\pm$  standard deviation (mean  $\pm$  SD).



Figure S6. Optical microscope images of 4T1 and HepG2 cells treated with CuFeSe<sub>2</sub>-PEG-FA and CuFeSe<sub>2</sub> (100  $\mu$ g/mL). The red arrows represent some targeting CuFeSe2-PEG-FA nanoparticles binding to the surface of 4T1 cells (b). The green arrows indicate some targeting CuFeSe<sub>2</sub>-PEG-FA nanoparticles binding to the surface of HepG2 cells (d). The yellow arrows represent some untargeted CuFeSe<sub>2</sub> nanoparticles binding to the surface of 4T1 cells (e). The blue arrows indicate some untargeted CuFeSe<sub>2</sub> nanoparticles binding to the surface of HepG2 cells (f). Scale bar: 10  $\mu$ m.



Figure S7 In vitro MR and CT imaging of FA blocking assay. (a) is the in vitro MR imaging; (c) is the MR signal intensity at different concentrations. (b) is the in vitro CT imaging; (d) is the CT value at different concentrations.



Figure S8 (a) and (b) are T2WI and CT images of 4T1 and HepG2 cells incubated with CuFeSe<sub>2</sub>-PEG-FA (200  $\mu$ g/mL, 2 mL), CuFeSe<sub>2</sub> (200  $\mu$ g/mL, 2 mL) and glucose solution (5%, 2 mL) for 4 h, respectively. (c) and (d) are the corresponding MR signal intensity and CT value respectively.



Figure S9 Fe contents in the tumors of targeted (CuFeSe<sub>2</sub>-PEG-FA), untargeted (CuFeSe<sub>2</sub>) and control group (5% glucose solution) by ICP-OES

Scanning	The T2* value of liver in	р	p The T2* value of liver $p$	
time	experiment group $(n = 3)$		in control group $(n = 2)$	
pre	5.68 ±2.50		$8.45 \pm 6.81$	
2 min	5.94 ±2.07	0.728	$8.83 \pm 6.18$	0.546
30 min	$5.40 \pm 1.62$	0.778	9.64 ±6.02	0.280
1 h	6.04 ±2.27	0.540	9.40 ±6.43	0.176
1.5 h	$6.47 \pm 2.76$	0.120	9.53 ±6.00	0.309
2 h	6.61 ±3.28	0.195	9.24 ±6.25	0.294
3 h	6.99 ±3.22	0.089	$9.17 \pm 5.89$	0.468
4 h	6.88 ±3.29	0.148	9.69 ±6.24	0.199
5 h	6.98 ±3.38	0.157	9.66 ± 6.17	0.228
6 h	6.97 ±3.03	0.115	$11.02 \pm 8.20$	0.232
8 h	7.15 ±2.53	0.730	9.67 ±6.17	0.212
12 h	6.80 ±3.16	0.168	$10.24 \pm 7.17$	0.090

Table S1. T2\* value of liver at different time points before and after tail vein injection

The T2\* values of liver before and after the injection were expressed as mean  $\pm$  SD.

Scanning	The T2* value of muscle in	р	The T2* value of muscle	р
time	experiment group (n = 3)		in control group $(n = 2)$	
pre	$15.04 \pm 6.60$		$18.14 \pm 2.20$	
2 min	14.73 ±4.72	0.904	$18.82 \pm 3.05$	0.461
30 min	$14.76 \pm 5.30$	0.897	$19.09 \pm 3.11$	0.378
1 h	$15.53 \pm 6.47$	0.711	$19.17 \pm 3.03$	0.328
1.5 h	$15.43 \pm 6.77$	0.840	$19.04 \pm 2.87$	0.308
2 h	$16.18 \pm 6.29$	0.569	$18.96 \pm 2.45$	0.133
3 h	$16.75 \pm 7.01$	0.371	$18.57 \pm 2.11$	0.094
4 h	$16.99 \pm 7.89$	0.442	$19.14 \pm 4.06$	0.585
5 h	$16.36 \pm 6.83$	0.493	$21.00 \pm 4.98$	0.383
6 h	$16.41 \pm 7.09$	0.435	$21.09 \pm 5.86$	0.459
8 h	$16.09 \pm 7.63$	0.605	$19.43 \pm 3.00$	0.262
12 h	$15.79 \pm 7.05$	0.724	$20.38 \pm 3.91$	0.315

Table S2. T2\* value of muscle at different time points before and after tail vein injection

The T2\* values of muscle before and after the injection were expressed as mean  $\pm$  SD.

Scanning time	CT value(HU)	р	Effective atomic number	р
	n=2		(n = 2)	
pre	43.45 <u>+</u> 4.45		7.40±0.04	
2 min	44.90±3.39	0.304	7.37±0.00	0.500
15 min	44.10±2.55	0.714	7.37±0.01	0.590
30min	$43.25 \pm 1.06$	0.967	7.40±0.04	0.942
45min	$45.05 \pm 1.20$	0.758	7.39±0.01	0.844
1h	$46.95 \pm 1.06$	0.383	7.36±0.02	0.205
1.5h	45.20±2.12	0.771	7.38±0.01	0.705
2 h	$44.15 \pm 1.34$	0.804	7.36±0.01	0.500
3 h	48.45±0.92	0.295	7.36±0.01	0.323
4 h	46.10±1.70	0.404	7.37±0.03	0.205
5 h	$46.85 \pm 1.48$	0.352	7.40±0.00	1.000
6 h	48.75±2.05	0.198	7.42±0.01	0.705
7 h	51.05±5.30	0.050	7.41±0.04	0.205
8h	52.50±3.82	0.032	7.37±0.03	0.205
12	50.95±4.74	0.017	7.37±0.04	0.205

Table S3. CT value and effective atomic number of tumors before and after intravenous injection.

CT value and effective atomic number of tumors before and after intravenous injection were expressed as mean  $\pm$ SD.