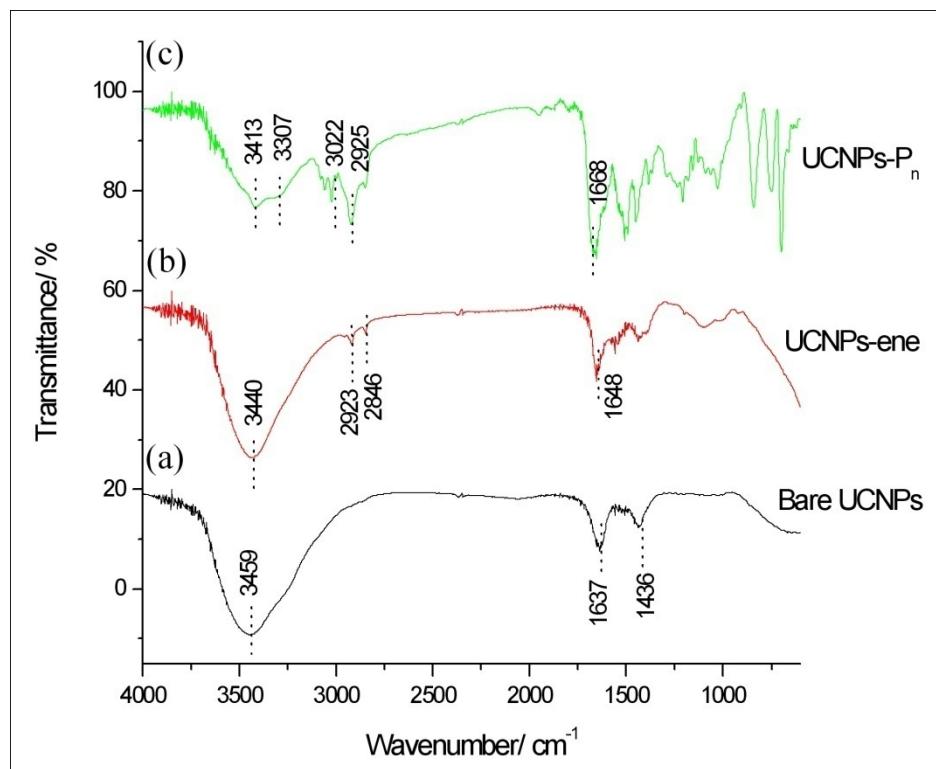
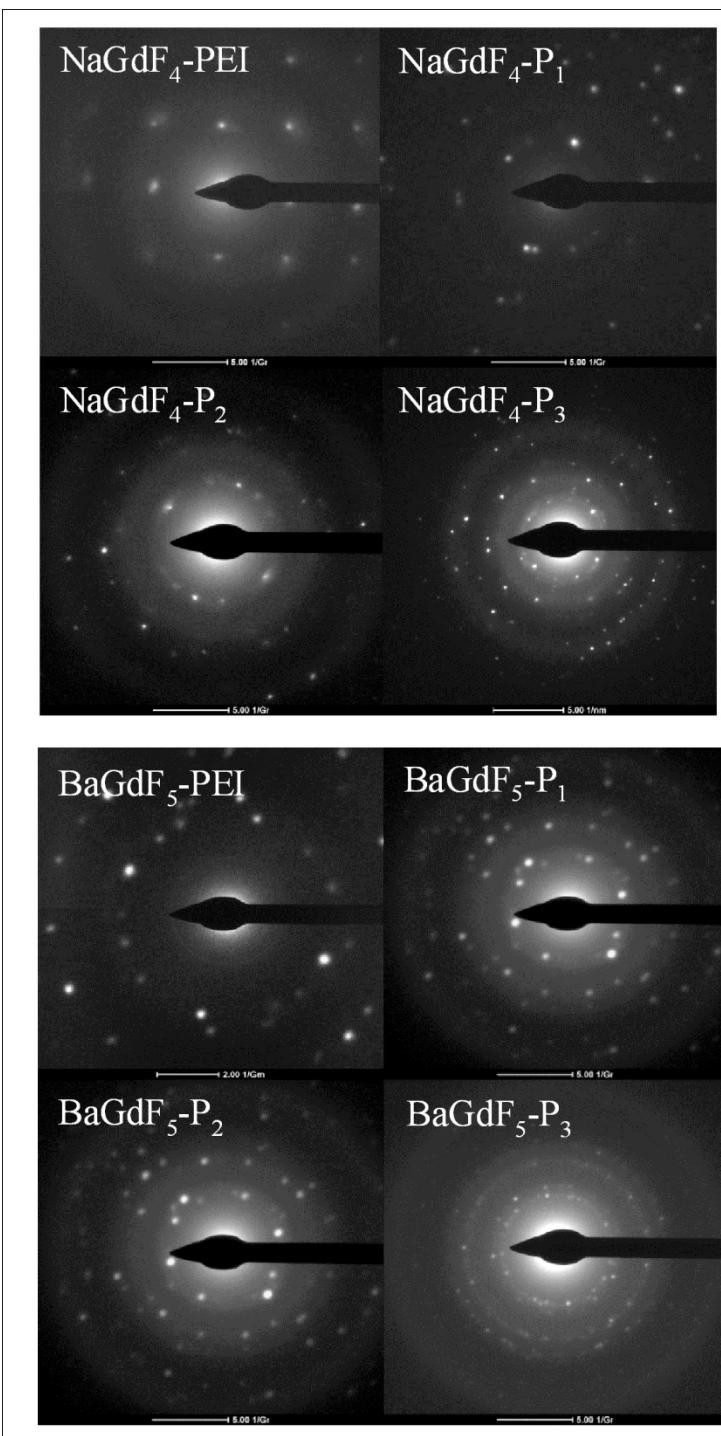


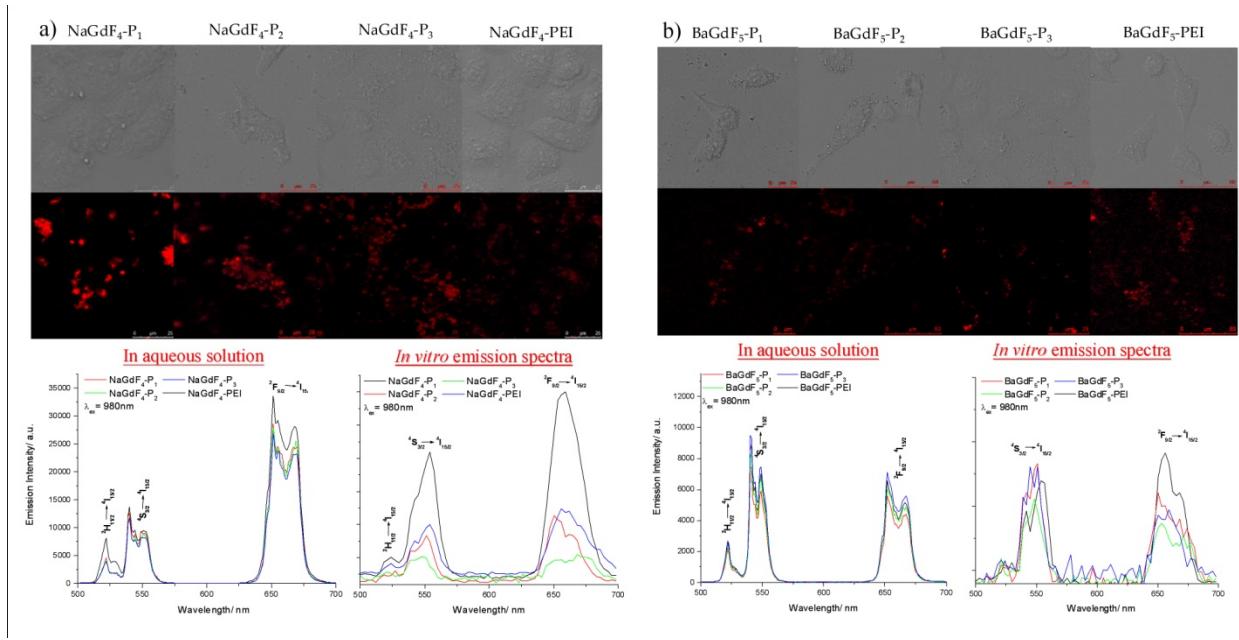
**Supporting Information**



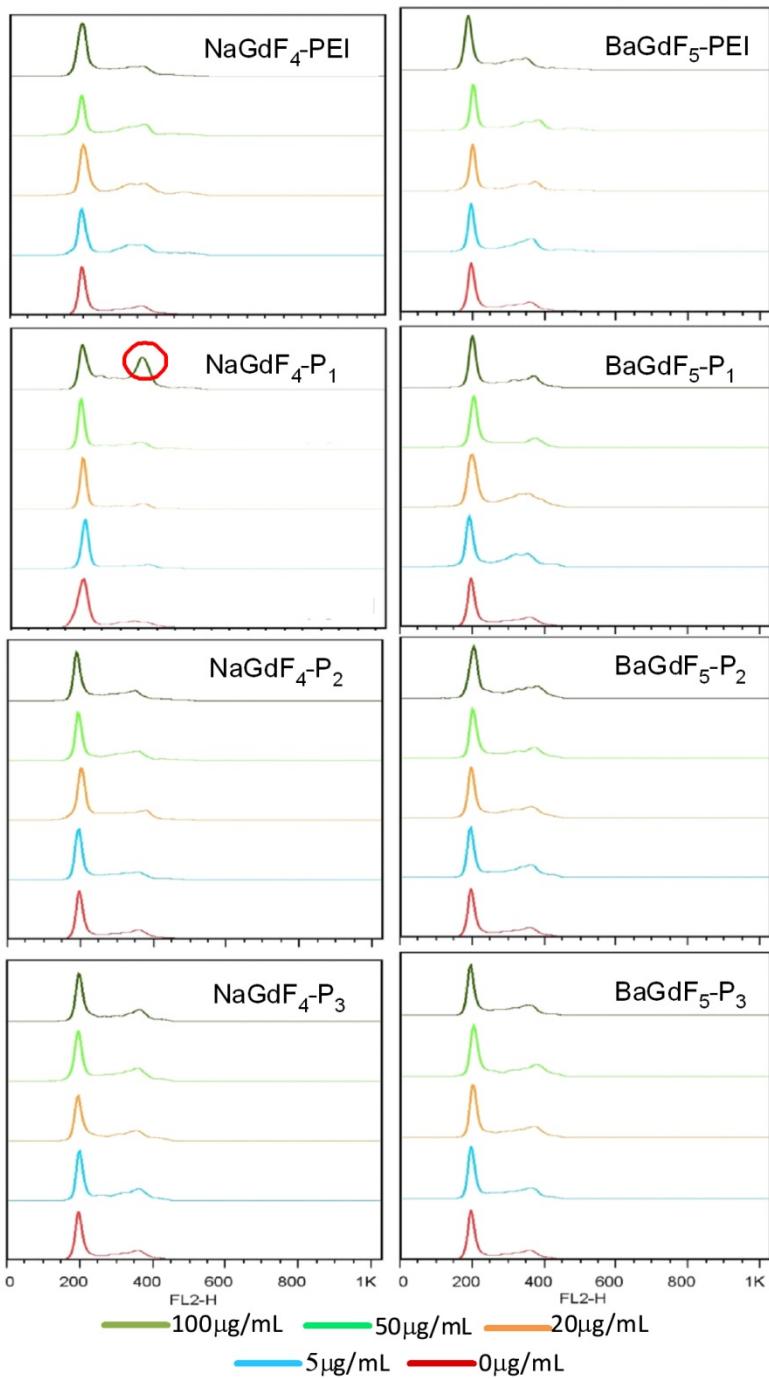
**Figure S1.** FTIR transmission spectra of (a) amine-functionalized **NaGdF<sub>4</sub>** and **BaGdF<sub>5</sub>** (UCNPs-PEI), (b) click reaction-modified **NaGdF<sub>4</sub>** and **BaGdF<sub>5</sub>** (UCNPs-ene) and (c) Plk1-specific peptides-coated **NaGdF<sub>4</sub>** and **BaGdF<sub>5</sub>** (UCNPs-P<sub>n</sub>).



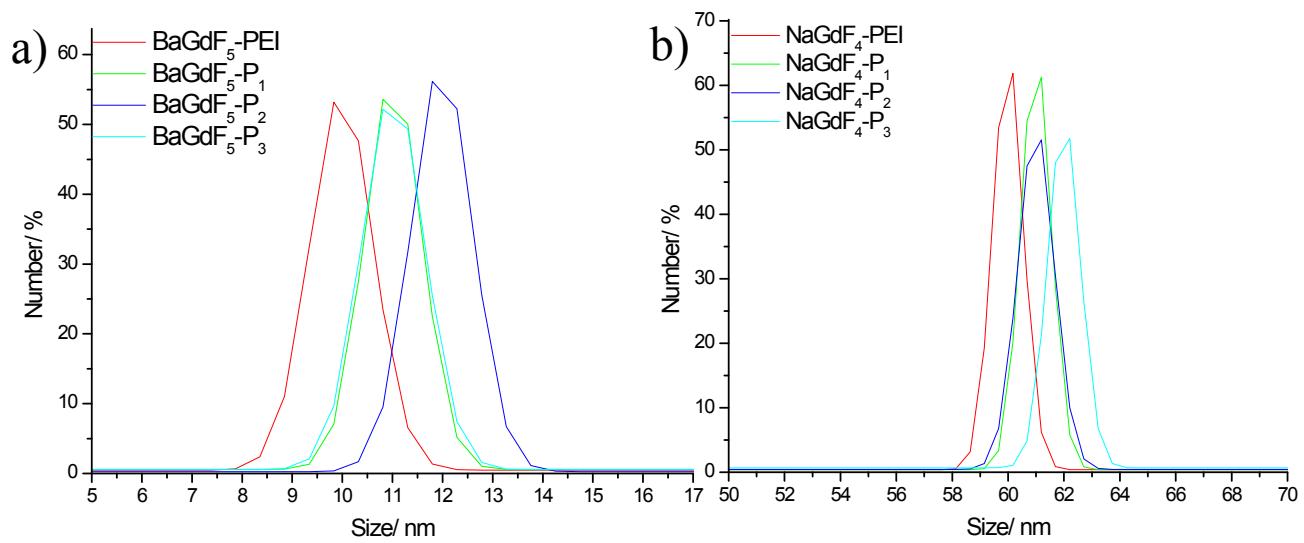
**Figure S2.** SAED patterns of **NaGdF<sub>4</sub>-P<sub>n</sub>** and **BaGdF<sub>5</sub>-P<sub>n</sub>**.



**Figure S3.** *In vitro* images and upconversion emission spectra in aqueous medium and *in vitro* of (a) **NaGdF<sub>4</sub>-P<sub>n</sub>** and (b) **BaGdF<sub>5</sub>-P<sub>n</sub>**.



**Figure S4.** Flow cytometric analysis of cell cycle interruption of HeLa under the treatment of  $\text{NaGdF}_4\text{-P}_n$  or  $\text{BaGdF}_5\text{-P}_n$ .



**Figure S5.** Dynamic light scattering analysis of a)  $\text{BaGdF}_5\text{-P}_n$  or b)  $\text{NaGdF}_4\text{-P}_n$  in PBS buffer, pH = 7.4.

$IC_{50}$ ( $\mu$ g/mL)	Cancer		Normal	
	HeLa	HK-1	MRC-5	QSG-7701
$NaGdF_4$ -PEI	398	516	435	432
$NaGdF_4$ -P <sub>1</sub>	103	223	334	401
$NaGdF_4$ -P <sub>2</sub>	111	183	383	609
$NaGdF_4$ -P <sub>3</sub>	95	172	400	732
$BaGdF_5$ -PEI	470	474	616	435
$BaGdF_5$ -P <sub>1</sub>	264	229	549	322
$BaGdF_5$ -P <sub>2</sub>	327	141	850	334
$BaGdF_5$ -P <sub>3</sub>	221	427	669	498

**Table S1.**  $IC_{50}$  values of the as-synthesized  $NaGdF_4$ -P<sub>n</sub> and  $BaGdF_5$ -P<sub>n</sub> in both cancer and normal cell lines.