

Figure S1. (a) TEM image and (b) UV-vis spectrum of GNRs.

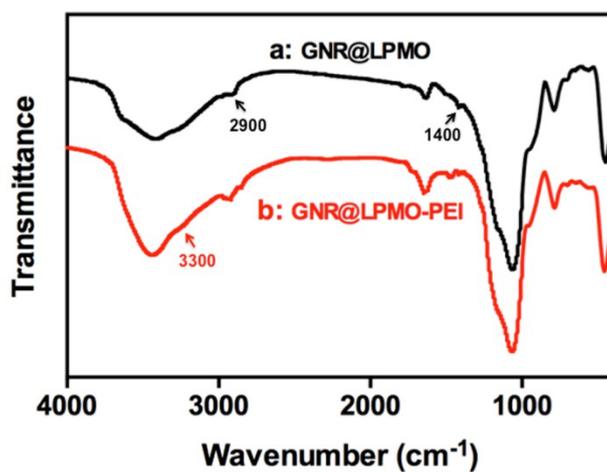


Figure S2. FTIR spectroscopy of the GNR@LPMO and GNR@LPMO-PEI nanoparticles.

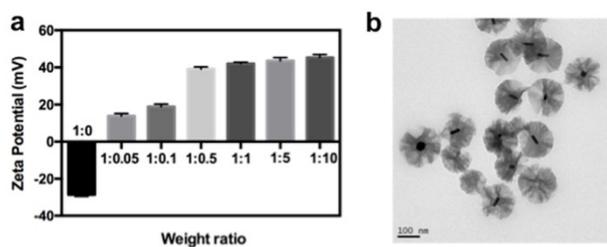
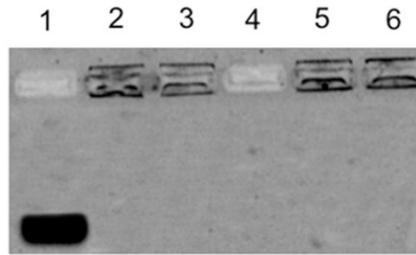
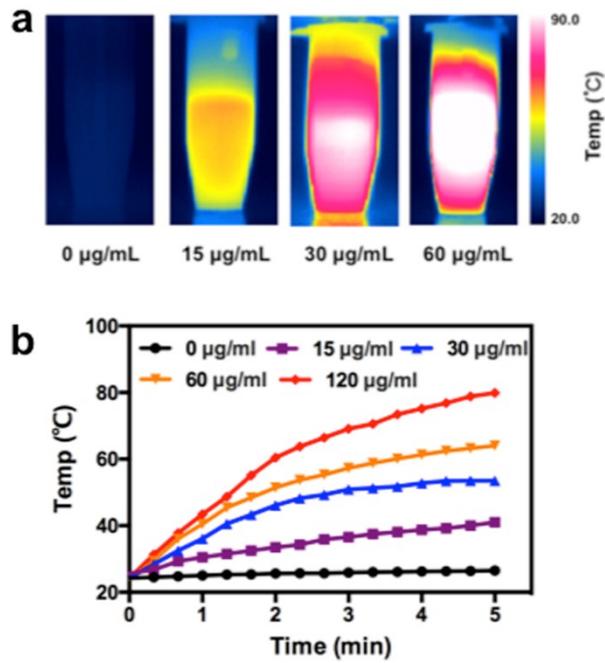


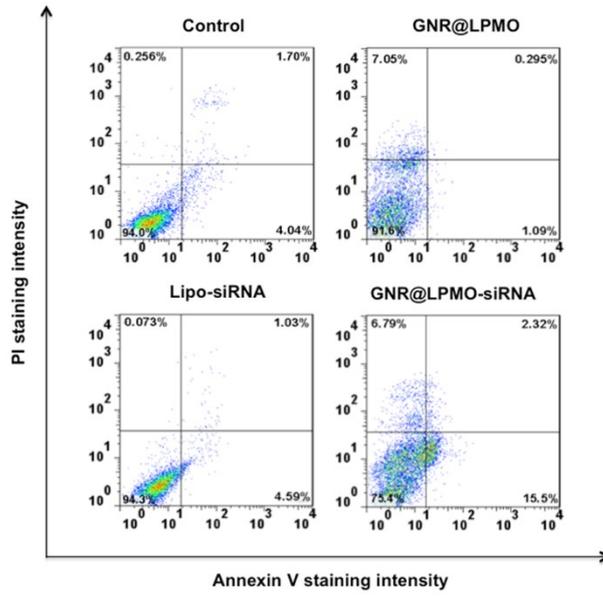
Figure S3. (a) Zeta potentials of the GNR@LPMO nanospheres coated with different amounts of PEI, (b) TEM image of GNR@LPMO-PEI nanospheres (weight ratio = 1:0.5).



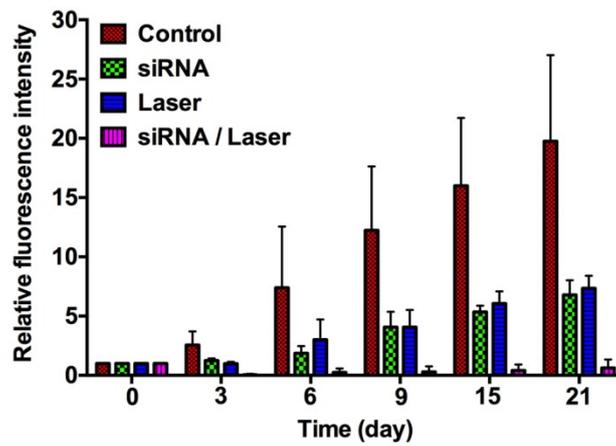
**Figure S4.** RNase protection assay. Lane 1: free siRNA; Lane 2-3: GNR@LPMO-siRNA complex nanoparticles with different weight ratio of 32, and 64; Lane 4: free siRNA + RNase A/T1; Lane 5-6: GNR@LPMO-siRNA + RNase A/T1, wight ratio = 32, and 64.



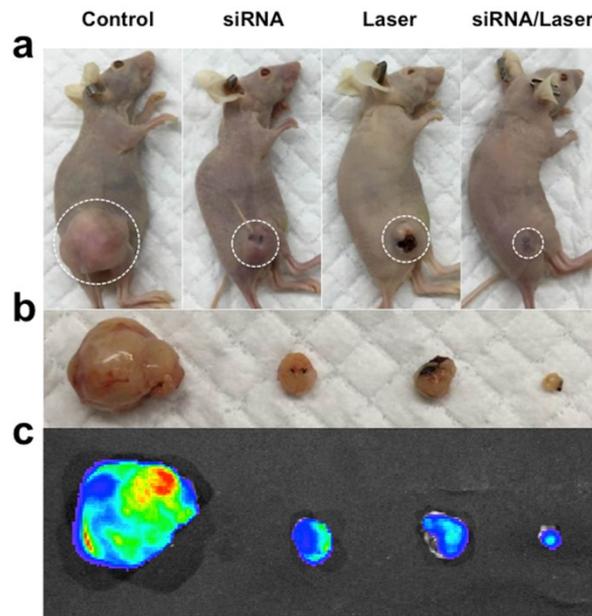
**Figure S5.** (a) Infrared thermal images of different concentrations of GNR@LPMO upon 808 nm laser irradiation (1.0 W/cm<sup>2</sup>, 5 min), (b) temperature increasing curves of different concentrations of GNR@LPMOs upon 808 nm laser irradiation in MEM solutions.



**Figure S6.** Cell apoptotic assay. Cell apoptosis was analyzed by flow cytometry in MDA-MB-231 LUC cells without treatment (control), treated with GNR@LPMO, Lipo-siRNA, GNR@LPMO-siRNA as external management.



**Figure S7.** Relative fluorescence intensity of MDA-MB-231 LUC tumor-bearing mice after different treatments.



**Figure S8.** Representative photos of MDA-MB-231 LUC tumor-bearing mice, and the resected tumors (bioluminescence) photographs (b, c) from different groups at 21 day post-treatment.