Supporting Information

Experimental Section

1. Materials and Apparatus.

1-Ethyl-3-(3-dimethyl aminopropyl) carbodiimide hydrochloride (EDC) and N-Hydroxy succinimide (NHS) were both obtained from Sigma-Aldrich. The deoxy-ribonucleoside triphosphate, Taq DNA Polymerase and 10×PCR buffer were all purchased from Shengon Biotechnology Co. Ltd. (Shanghai, China). Water used in the whole experiment was deionized and purified to 18.2 MW (Millipore). All the DNA fragments were synthesized by Shengon Biotechnology Co. Ltd. Their sequences are listed as follow:

Target DNA:

5'-GAAGGACGAAGGACTCTAACGTTTAACATCCTTTGCCATTG-3'

Upstream primer: 5'-GAAGGACGAAGGACTCTAACG-NH_2-3'

Downstream primer: 5'-CAATGGCAAAGGATGTTAAACG- NH_2-3'

Nonspecific DNA:

5'-GATCGGGTGTGGGTGGCGTAAAGGGAGCATCGGACAATCAA-3'

PCR was performed by an eppendorf PCR-authorized thermal cycler (eppendorf, USA). Transmission electron microscopy (TEM) images were obtained using a JEOL 2100 microscope (Japan). The size distribution of the MNPs assembly was measured by a dynamic light scattering instrument (Malvern Zetasizernano, England). T2 was determined on the Niumag-NMI20-Analyst (Shanghai Niu-mag Corp.) and the MRI images were obtained on the MiniMR-60 (Shanghai Niu-mag Corp.), the

field intensity were 0.5T and 0.55T respectively.

2. The experiment parameters

The PCR mixture was reacted in a final volume of 50 μL, which consisted of 2 μL of upstream and downstream primers modified MNPs respectively, 5 μL of 10×PCR buffer, 1 μL of 10 mM dNTP, 0.5 μL Taq DNA polymerase, 1 μL of DNA template with different concentrations from 0.01 fM to 10000 fM and 39.5μL of distilled H_2O. The PCR parameters: an initial denaturation 2 min at 94 °C, followed by 30 cycles at 94 °C for 30s, 60 °C for 30s, 72 °C for 1 min, and a final extending 7 min at 72 °C.

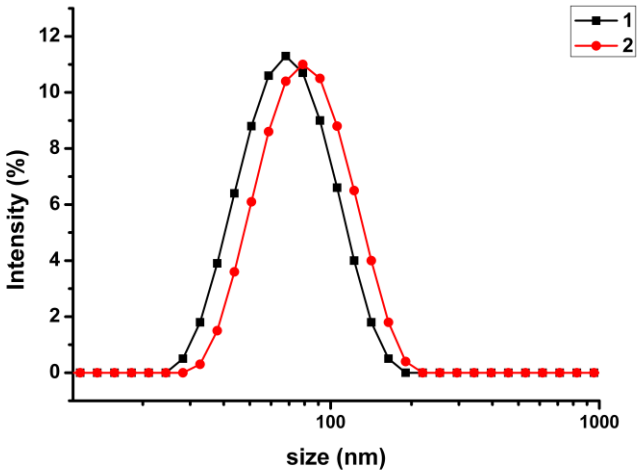

Figure S1. The size change of magnetic nanoparticles before and after DNA conjugation. 1: the original MNPs, 2: the MNPs modified with DNA.

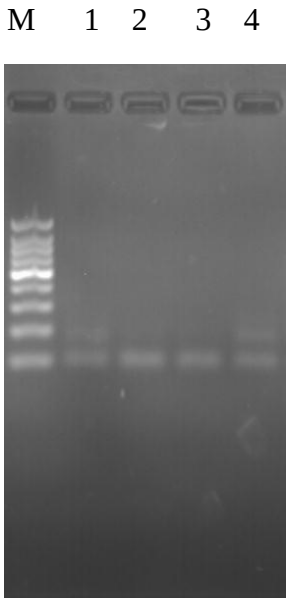

Figure S2. 2.5% agarose gel analysis of PCR products. M: 50bp DNA marker. Lanes 1-4: the annealing temperatures were 55 °C, 58 °C, 60 °C, 65 °C, respectively.

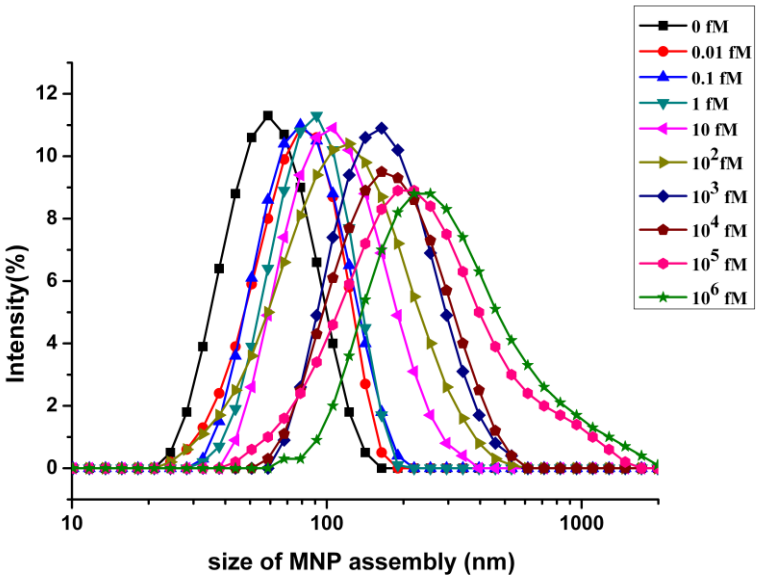

Figure S3. Typical size distributions of the assembly of MNPs under different concentrations of target DNA in the range of 0.01 fM to 10000 fM.

Table S1. Determination of target DNA spiked into λDNA.

Spiked concentration (fM)	Detected concentration Mean[a]±SD[b] (fM)	Recovery (%)
50	48.42±2.421	96.8
10	10.30±0.515	103
5	4.75±0.2375	95
1	0.92±0.046	92
0.5	0.489±0.02445	97.8
0.1	0.096±0.0048	96
0.05	0.053±0.00265	106

a. The mean of three experiments.
b. SD=standard deviation.