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Supporting Information

Experimental Section

Reagents and Materials:

DNA sequences used in the study:

Target DNA: 5'-CACGCACACACACTTTAAA-3'

Second target: 5'-GGGTTTTGGGTTTTGGGTTTTGGGTTTTCACGCACAGTCACCTAA-3'

Hairpin probe:

All DNA were synthesized by Shanghai Sangon Biological Engineering Technology & Services (Shanghai, China). Exonuclease Ⅲ (Exo Ⅲ) was purchased from Takara Biotechnology Co. Ltd. (Dalian, China) and used without further purification. NMM was purchased from Porphyrin Products (Logan, UT). Other chemicals were purchased from Sigma-Aldrich and used without further purification. *E.coli* (ATCC 25922) and *S.aureus* (ATCC 25923) bacterial strains were obtained from Chuanxiang Biotechnology, Ltd. (Shanghai, China). Ultrapure water (18.2 MΩ; Millpore Co. USA) was used throughout the experiment.

Apparatus and characterization: UV-Vis spectroscopy spectra were recorded on a Cary 300 UV-Vis spectrophotometer. Steady-state fluorescence measurements and time curves were carried out by using a JASCO FP-6500 spectrofluorometer with a temperature-controlled water bath. Fourier transform infrared spectroscopy (FT-IR) measurements were carried out on a Bruker Vertex 70 FITR spectrometer. Scanning electron microscopic (SEM) images were recorded using a Hitachi S-4800 Instrument (Japan). Transmission electron microscopy (TEM) images were recorded using a FEI TECNAI G220 high-resolution transmission electron microscope operating at 200 kV. The zeta potential of the nanoparticles in water was measured in a Zetasizer 3000HS analyser. Dynamic Light Scatterer (DLS) was made by Malvern Corp, U. K. (ZEN3690). XPS measurement was performed on an ESCALAB-MKII spectrometer (VG Co., United Kingdom) with Al KR X-ray radiation as the X-ray source for excitation.

Preparation of superparamagnetic Fe_3O_4 nanoparticles: Fe_3O_4 nanoparticles were synthesized according to the literature.[1] Firstly EN (8.9 mL) was mixed with EG (80 mL), then $FeCl_3 \cdot 6H_2O$ (2.50 g) was added under magnetic stirring, followed by the addition of NaAc (5.00g). After that, the mixture was heated at 200 °C for 6 h after transferring to a Teflon-lined stainless-steel autoclave. The produced magnetite nanoparticles were separated using a magnet, and then washed with deionized water. Finally the production was dried in an oven at 80 °C for 12 h.

Preparation of Fe_3O_4@SiO_2 nanoparticles: Fe_3O_4@SiO_2 nanoparticles were prepared according to a previously reported method.[2] the magnetite particles (0.1g) were thoroughly washed with deionized water and homogeneously redispersed in a solution containing ethanol and deionized water (4:1), and then concentrated ammonia aqueous solution (1.0 mL, 28 wt.%) was added with continuously ultrasonication. Next, TEOS (1.0 mL) was added dropwise into the above solution slowly. Following that the mixture was allowed to react for 2 h and then transferred to take a mechanical stir for 6 h. Then Fe_3O_4@SiO_2 nanoparticles were collected via an assistant magnet and washed with deionized water and ethanol in sequence.

Preparation of QMNPs nanoparticles: Fe_3O_4@SiO_2 nanoparticles (25 mg) were homogeneously dispersed in isopropyl alcohol solution (60 mL), and then TMSPTMAC (200 μL) was added slowly with the help of ultrasonication. Following that the mixture was allowed to react for 4 h. The liquid was removed, and the QMNPs were stored in a vacuum oven for 48 h at 140 °C, washed with water several times under magnet, and then dried at 80 °C.

Preparation of DNA-QMNPs nanoparticles: The QMNPs nanoparticles were dispersed into phosphate buffer saline (PBS) solution (400 μL) with the concentration of 0.8 g L^{-1}. And then 1.5 μL target DNA (357.8 μmol L^{-1}) was added into above solution, followed by shaking for 20 min so as to ensure reaching equilibration. After magnetic separation, we obtained the solid of DNA-QMNPs nanoparticles. The supernatant was collected to analyze the adsorption quantity. The adsorption curves were obtained by measuring the solution absorbance at 260 nm before and after the treatment using UV-Vis spectroscopy.

Gel electrophoresis: Firstly, all DNA samples were annealed. Then, samples needed hydrolysis were initiated by addition of Exo III and maintained at 37 °C (reaction mixtures contained 50 U of Exo III). After 40 min, digestions were stopped by adding stop buffer. Finally, gel electrophoresis was carried out on polyacrylamide gel (15 %, W/V) with 29:1 ratio of acrylamide: bisacrylamide, at 25 °C, 1 × TB buffer. Gel was silver stained. [DNA]= 3 μM.

Exo III-assisted DNA amplification assay: Firstly, the hairpin DNA probe was incubated with different concentrations of target DNA, followed with 50 U Exo Ⅲ, The mixture was equilibrated for 40 min at 37 °C. Finally, NMM was added and equilibrated for 5 min before spectral measurements. The final concentrations of hairpin probe and NMM were 1 μM and 1 μM, respectively. In fluorescence measurements, the samples were excited at 399 nm, and emission spectra were collected from 520 to 720 nm. All measurements were performed in phosphate buffer saline (PBS) containing 100 mM KCl, 10 mM $MgCl_2$, pH = 7.2.

Bacterial culture: Monocolony of *E.coli* and *S.aureus* on the solid Luria-Bertani (LB) agar plate was transferred to 20 mL of liquid LB culture medium and grown at 37 °C for 12 h under 170 rpm rotation. Then the resultant bacterial cells were separated by centrifugation (5000 rpm). Next, the bacterial cells were rinsed with PBS buffer and designed bacterial concentrations were adjusted by measuring the optical density at 600 nm (OD_{600}).

The effect of QMNPs concentration for DNA-capture ability: The concentration of QMNPs used for the DNA adsorption played a role in the final signal obtained. At first, the concentration of DNA was defined with 1.5 μM. Next a series of QMNPs concentration were chosen to mix with DNA solution for 1 h. Finally solution was separated by a magnet and the supernatant was taken for determination by UV-Vis spectrum at 260 nm.

Bacterial detection using Exo III-assisted DNA amplification assay: Firstly, the DNA-QMNPs nanoparticles were dispersed into PBS solution (400 μL) which contains bacteria with different concentration. And then, the solution was kept in shaking for 20 min, which would result in the complete interaction between bacteria and DNA-QMNPs nanoparticles. After that, the solution was separated by a magnet and 200 μL of the supernatant was transferred into the 200 μL solution containing the mixture of hairpin DNA probe and Exo III. Finally, DNA amplification assays were carried out.

Anti-interference Experiments: Firstly, the interference of bacterial secretion was carried out. The as-prepared bacteria solution (OD_{600}= 2.0) was centrifuged at 3000 rpm for 5 min and then the supernatant (400 μL) was mixed with DNA-QMNPs for 20 min. Then, magnetic separation was worked and 200 μL of the supernatant was transferred into the 200 μL of the mixture of hairpin DNA probe and Exo III. Finally, the above solution was detected by spectrofluorometer. Secondly, the control of bacterial secretion was set up. After disrupting the cells (OD_{600} = 2.0) by sonication on the ice, the bacterial extraction was obtained by centrifugation at 4 °C with 12,000 rpm for 20 min. The next steps were undertaken as above process.

Supporting Figures

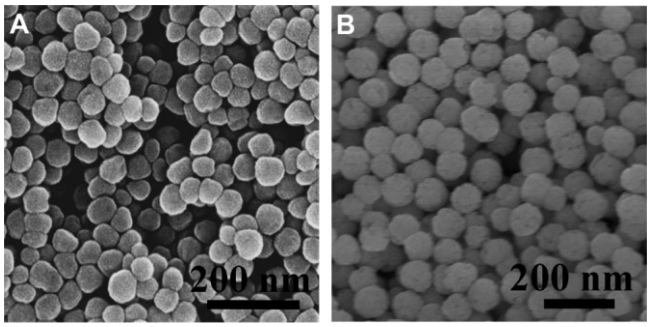

Fig. S1 (A) SEM micrograph of Fe_3O_4. (B) SEM micrograph of Fe_3O_4@SiO_2.

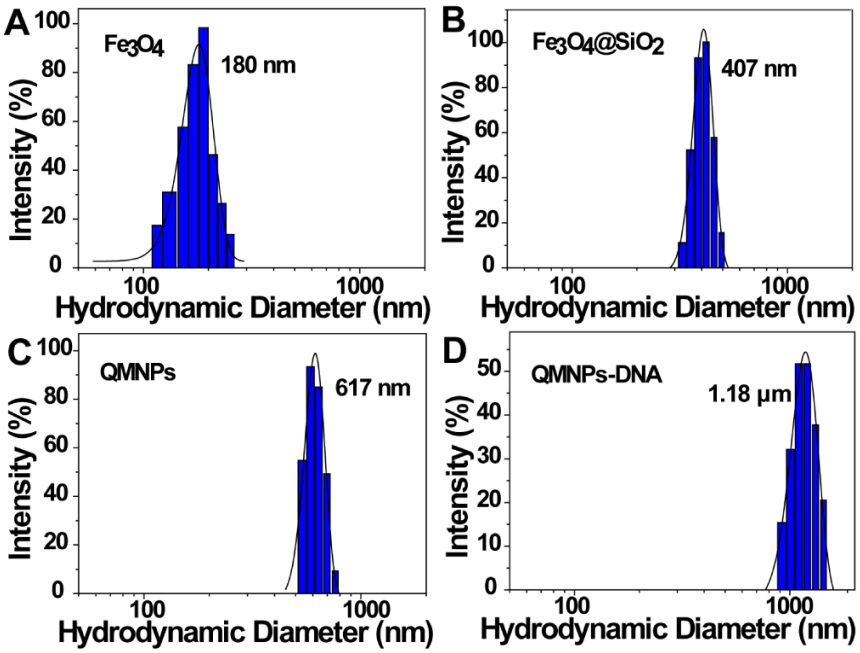

Fig. S2 The hydrodynamic diameter of (A) Fe_3O_4, (B) Fe_3O_4@SiO_2, (C) QMNPs and (D) QMNPs-DNA measured in water.

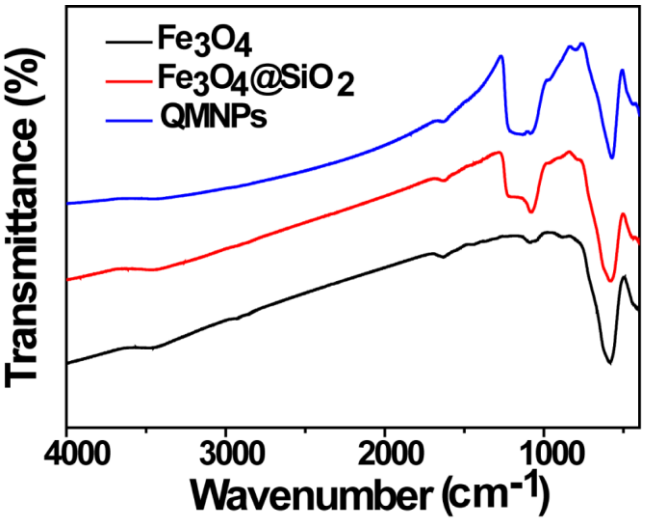

Fig. S3 Fourier transform infrared (FT-IR) spectra of Fe_3O_4, $Fe_3O_4@SiO_2$ and QMNPs.

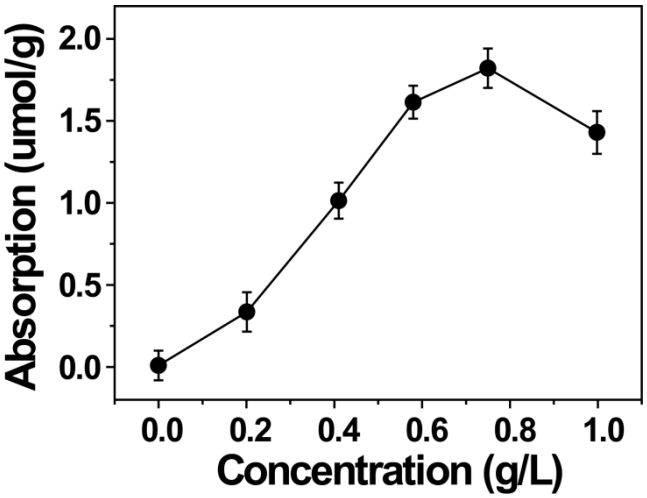

Fig. S4 The study of DNA-capture ability of QMNPs with the different concentrations of QMNPs. UV-Vis spectroscopy spectra at 260 nm. [DNA] = 1.5 μmol L^{-1}.

$$\text{Absorption} = \frac{A_{before} - A_{after}}{\varepsilon\, b\, c_{QMNPs}}$$

A_{before} means the UV absorption of origin DNA solution. A_{after} equals the UV absorption of supernatant of the blend of DNA and QMNPs after magnetic separation.

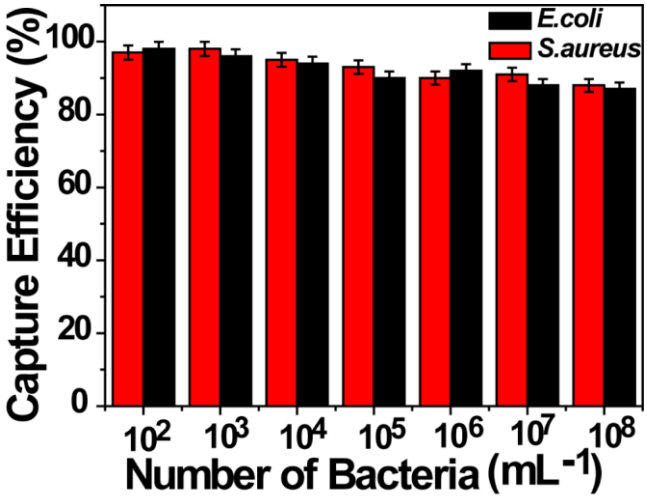

Fig. S5 Bacteria-capture ability of the QMNPs at different bacteria species and numbers. The error bars represent the standard deviation of three measurements.

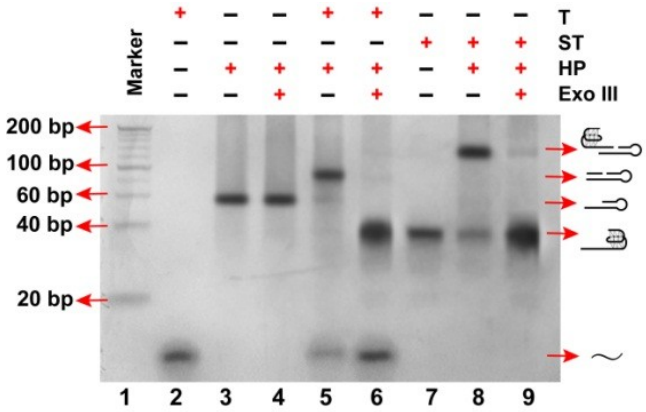

Fig. S6 Native gel electrophoretic analysis of the Exo III-assisted amplification strategy for DNA. Lane 1, DNA marker; Lane 2, T (T = target DNA); Lane 3, HP (HP = hairpin probe); Lane 4, HP + Exo III; Lane 5, T + HP; Lane 6, T + HP+ Exo III; Lane 7 ST (ST = second target); Lane 8, ST + HP; Lane 9, ST + HP + Exo III. [DNA] = 3 μM

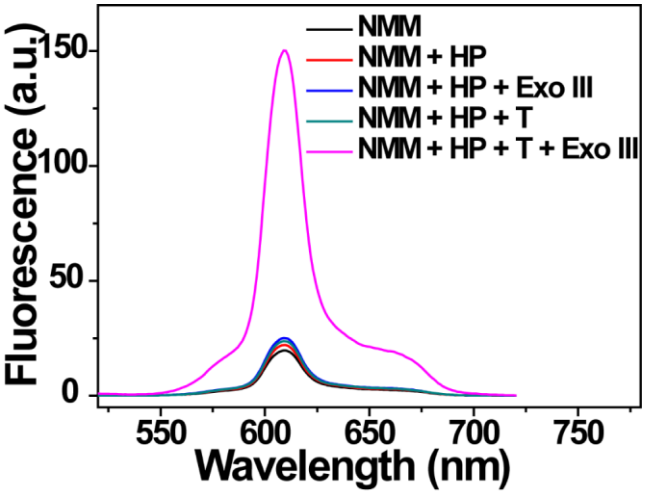

Fig. S7 Target DNA triggered Exo III to digest the hairpin probe and brought about the fluorescence enhancement. Control samples were indicated in the figure. [NMM]= 1 μM, [HP] = 1 μM, [T] = 2.5 nM.

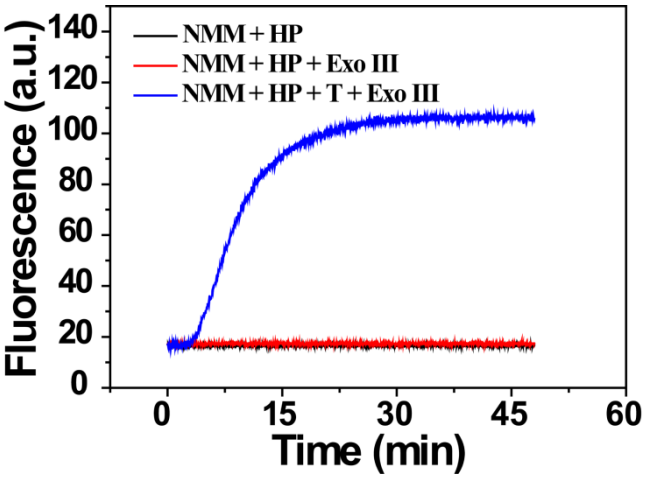

Fig. S8 Significant enhancement of fluorescence signal triggered by target DNA with time elapsing. [NMM] = 1 μM. [HP] = 1 μM. [T] = 1.5 nM.

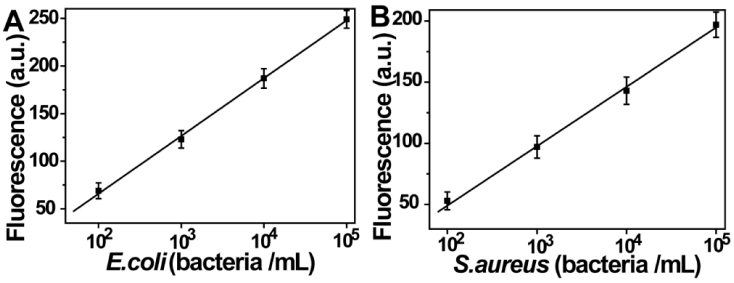

Fig. S9 Linear standard curves from 50 to 1 × 10^5 mL-1 of *E.coli* (A) and 80 to 1 × 10^5 mL-1 of *S. aureus* (B) were obtained during the detection process of bacteria, respectively.

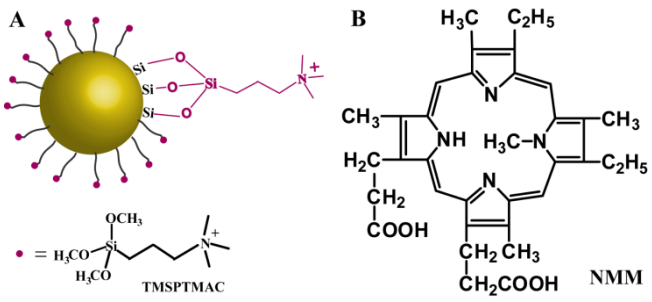

Fig. S10 (A) The diagram of QMNPs (QMNPs = Fe_3O_4@SiO_2@TMSPTMAC) and the structure of TMSPTMAC. (B) The molecular structure of NMM.

Method	Components	Bacterial strain	Detect limit (CFU/mL)	Specificity	Advantages	Challenges	Ref
standard plate count	agar plates	aerobic bacteria		-	Broad linearity range; good repeatability	Lengthy time; skilled personnel; low efficiency	[2]
polymerase chain reaction (PCR)	polymerase PCR primer	*E. coli* *Salmonella spp. B. cereus*	1	-	Highly specificity	Time consuming (12h); skilled personnel; high cost	[3]
ELISA	Antibody, polystyrene microsphere	*E. coli O157*	10^2	+	Highly specificity	trained operators; high-cost biomaterials	[4]
electrochemical methods	TiO_2 nanowire	*Listeria*	10^2	-	Rapid response, highly sensitive	Skilled laboratory personnel	[5]
colorimetric strategies	DNAzyme, urease	*E. coli*	500	+	Superior specificity; naked-eye detection	Complicated combination between DNA and urease.	[6]
	AuNPs, β-galactosidase		10^2	-			[7]
Luminescent	HSA-AuNCs	*S. aureus MRSA*	10^6	+	selective	sensitivity	[8]
Dark-field imaging	AuNPs, antibodies	*E. coli*	10^4	+	simple and rapid	Difficult antibodies modification	[9]
SERS	AgNPs, 4-mercaptophenylboronic acid	*E. coli* *S.aureus*	10^2	+	Sensitive and rapid	Strict requirement for surface of specific metal	[10]
fluorescent assay	carbohydrate-functionalized fluorescent polymer	*E. coli*	10^4	-	simple, rapid, and ultrasensitive	Complicated chemical modification	[11]
	Magnetofluorescent nanoparticles; vancomycin	*S. aureus*	1300				[12]
MALDI-MS	magnetic nanoparticles	*S. aureus*	10^4	+	Simple operation	Specific and expensive instrument	[13]
DNA amplification detection	QMNPs, and DNA probe	*E.coli*	50	-	Rapid, simple and sensitive	Lack of specificity	Present assay
		S.aureus	80				

Table S1 Comparison of the representative assays for the detection of bacteria.

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