Supporting information

Experimental Section

Reagents and equipments. All DNA sequences were all synthesized by Sangon (Shanghai, China). Tris-(2-carbozyethyl) phosphine hydrochloride (TCEP) was purchased from Sigma-Aldrich (St. Louis, MO) and used without further purification. Mercaptoethanol (MCE) was purchased from Fluka and used as received. All aqueous solutions were prepared using ultra-pure water (18.2 MΩ, Milli-Q, Millipore).

The metallo-supramolecular cylinder $[Fe_2L_3]Cl_4$ was synthesized and purified as previously reported. The enantiomerically pure $[Fe_2L_3]Cl_4$ was obtained by using a cellulose (~20 μm, Aldrich) column and eluting with 20mM NaCl aqueous solution.The purity is more than 95%, which was determined by ESI-MS and elemental analysis. UV-vis spectroscopy was used to determine the enantiomer concentration. The samples of purified P- and M-enantiomer were collected and freeze-dried for future use.

Electrode Cleaning and E-DNA Sensor Preparation. The DNA modified sensor was fabricated by using gold disk electrodes (Φ= 2 mm, CH Instruments, Austin, TX). The electrodes were prepared by polishing with 0.3 and 0.05 μm deagglomerated γ alumina (BUEHLER, UAS) suspensions followed by sonication in water and multiple steps of electrochemical cleaning before modification with the thiolated probe DNA. The clean gold surface was incubated with a 0.1 μM solution of thiolated DNA oligomer pretreated with TCEP in buffer (10 mM Tris buffer, 1.5M NaCl, 10m M $MgCl_2$, pH 7.2) for 12 h at room temperature. The surface was then rinsed with buffer and subsequently passivated with MCE in the Tris buffer for 1 h. Then, the electrodes were rinsed again with distilled water prior to measurements.

Electrochemical Measurements. Alternating current voltammetry (ACV) were performed with a CH Instruments model 660B electrostation (Austin, TX) in a standard cell with a platinum counter

electrode and an Ag/AgCl reference electrode. The E-DNA sensor measurements were conducted by monitoring the modified working electrode in Tris buffer (89 mM Tris salt, 89 mM borate, 10 mM NaCl, pH 7.2) using ACV with a step potential of 10 mV, amplitude of 25 mV, and a frequency of 10 Hz. Electrochemical impedance spectroscopy (EIS) was performed using Solartron Instrument equipped with S11287 electrochemical interface and S11255 HF Frequency response analyzer in buffer (89 mM Tris salt, 89 mM borate, 10 mM NaCl, pH 7.2) as the supporting electrolyte. The impedance spectra were recorded at potential of 0.165V within the frequency range of 10^{-2}-10^{5} Hz. The amplitude of the applied sine wave potential in each case was 5 mV.

For chiral complexes detection measurements, compounds were diluted with the aqueous Tris buffer, then the modified electrodes were incubated in each sample containing different concentrations of complexes and MB-labeled DNA for 20min at room temperature except the time-course study. Prior to measurement, washing with the measured buffer is necessary. The regeneration was done with a simple 30s hot ultrapure water (50℃) rinse.

Figure S1: Structures of the M-enantiomer (left) and P-enantiomer (right) of $[Fe_2L_3]^{4+}$ cation (up). Iron: yellow; carbon and nitrogen atoms in three ligands L are shown in red, green and blue, respectively. Hydrogen atoms are omitted for clarity. The crystal data of $[Fe_2L_3]^{4+}$ are from the Cambridge Crystallographic Data Centre CCDC 622770. The structure of bis(pyridylimine) ligand L (down).

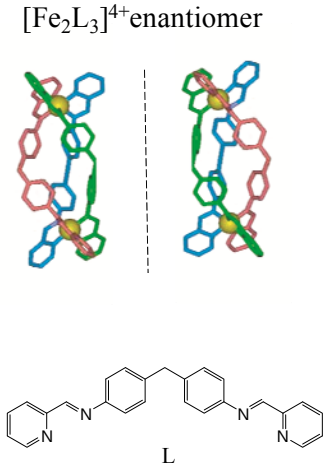


Figure S2: Time-dependent curve of then E-DNA sensor.

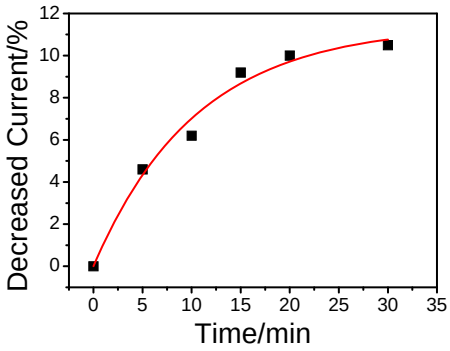


Figure S3: Optimal frequency of the E-DNA sensor.

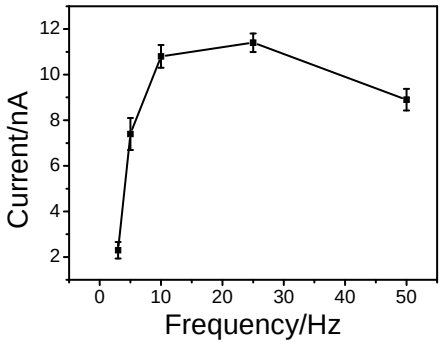
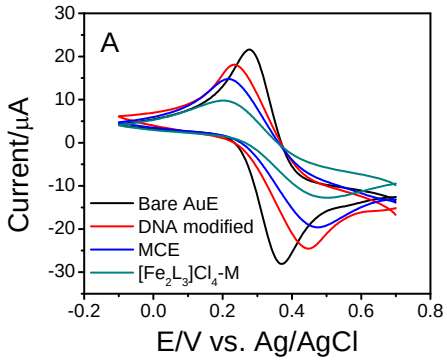
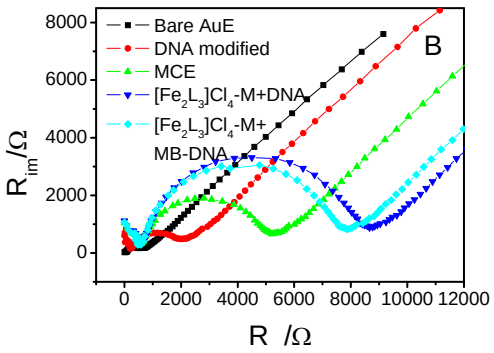


Figure S4: CVs and EIS of the modified electrode.





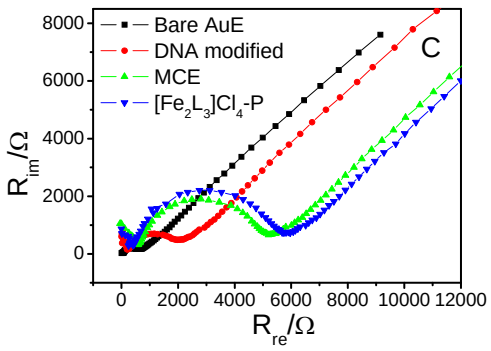


Figure S5: The linear relationship between the current and the concentration of $[Fe_2L_3]Cl_4$-M/-P.

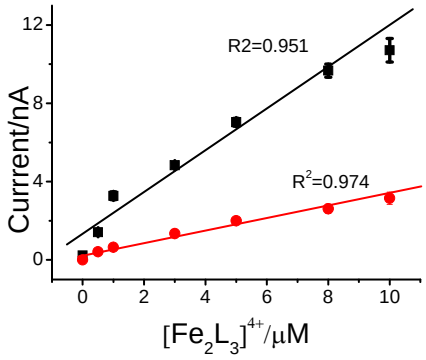


Figure S6: UV-melting curves of DNA, DNA with $[Fe_2L_3]Cl_4$-M/-P.