Supplementary Information

A fluorescence and surface-enhanced Raman scattering dual-mode aptasensor for rapid and sensitive detection of lead ion

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Table S1 DNA Sequences used in this work

Note	Sequence
M-PS2.M	5′-GAT ATC AGC GAT TTT TGT GGG TAG GGC GGG TTG GAA AAC ACC CAT GTT ACT CT-3′
H1	5′-GTT CGT CAA AAA GAG TAT /rA/GG ATA TCG CGT GAC GAA CTA GTT GAT GAA GCT G-SH-3′
A1	5′-Cy3-GTG TGC CTA TTA TGT CTC CTC CTG TGT GCC TAT TAT GTC TCC TCC TCA GCT TCA TCA ACT AGT TCG TCA TTT TT-SH-3′
A2	5′-AAC TAG TTG ATG AAG CTG GAC ATA ATA GGC ACA CGA CAT AAT AGG CAC AC-3′
Assistant A	5′-GTG CCT ATT ATG TCG TGT GCC TAT TAT GTC CAG CTT-3′
B1	5′-Cy3-AGG AGG AGA CAT AAT AGG CAC ACT GAC GAA CTA GTT GAT GAA GCT GTT TTT-SH-3′
B2	5′-CAG CTT CAT CAA CTA GGT GTG CCT ATT ATG TCT C- 3′
Assistant B	5′-GCA CAC CTA GTT GAT GAA GC-3′
Trigger DNA	5′-GGA TAT CGC GTG ACG AAC TAG TTG ATG AAG CTG-3′

Optimization of method

To achieve optimal sensing performance, different experimental parameters were investigated at a Pb^{2+} concentration of 100 pM and other experimental conditions were the same.

In Fig. S1, the cleaving activity of M-PS2.M initially increased and then decreased with increasing pH, corresponding to the changing trend of FL-SERS signal. The cleaving activity of M-PS2.M reached its maximum at pH 7.4. Therefore, pH 7.4 was chosen as the optimum condition. In Fig. S2, the changes in FL-SERS intensity during different time intervals of the cleaving reaction were shown. The FL signal intensity gradually decreased with time while the SERS signal gradually increased until reaching their minimum and maximum values at 60 mins, separately. Beyond 60 mins, the signal intensities tended to stabilize, thus a cleaving time of 60 mins was selected. In Fig.S3, the changes in FL-SERS intensity resulting from the cleaving activity of M-PS2.M at different reaction temperatures were presented. It can be observed that both minimum FL signal intensity and maximum SERS signal intensity were generated at 37 °C. Hence, 37 °C was chosen as the optimal temperature for the cleaving reaction.

In Fig.S4, during the nonlinear HCR nanoassembly process, the most ideal FL-SERS signal intensity was achieved when substrate A and substrate B were in a ratio of 1:2. This is because the ratio of substrate A to substrate B affects the degree of nonlinear HCR reaction. In Fig.S5, the effects of different reaction times on the degree of

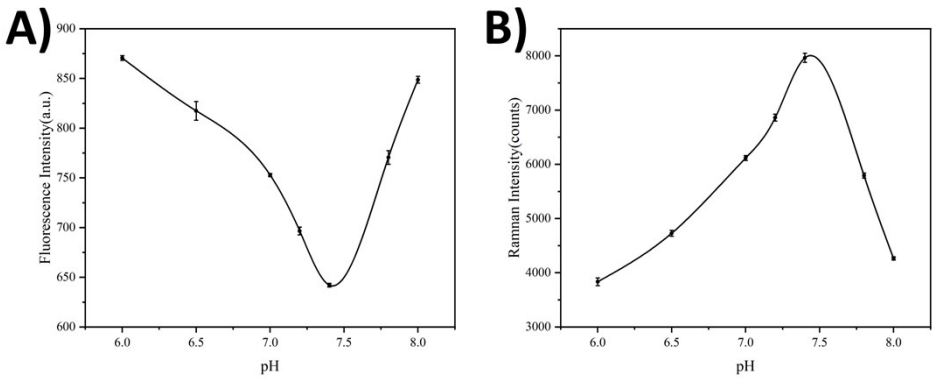

Fig. S1 Effect of pH on the (A) FL intensity and (B) SERS intensity for 100 pM Pb^{2+}.

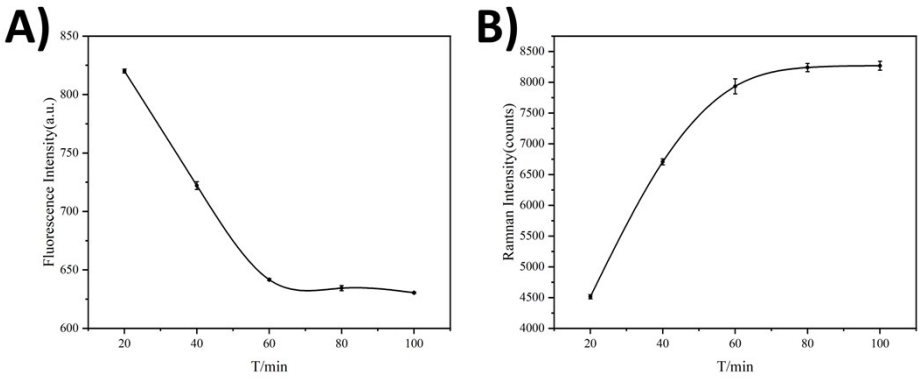

Fig. S2 Effect of cleaving time on the (A) FL intensity and (B) SERS intensity for 100 pM Pb^{2+}.

nonlinear HCR were studied. With increasing time, the FL signal gradually diminished while the SERS signal increased, reaching their respective minimum and maximum values at 90 mins. Beyond 90 mins, the signals tended to stabilize. Therefore, a nonlinear HCR nanoassembly time of 90 mins was selected.

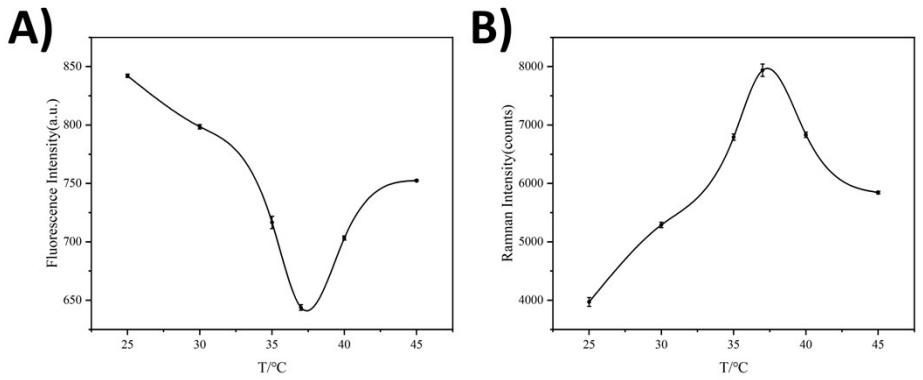

Fig. S3 Effect of the cleaving temperature on the (A) FL intensity and (B) SERS intensity for 100 pM Pb^{2+}.

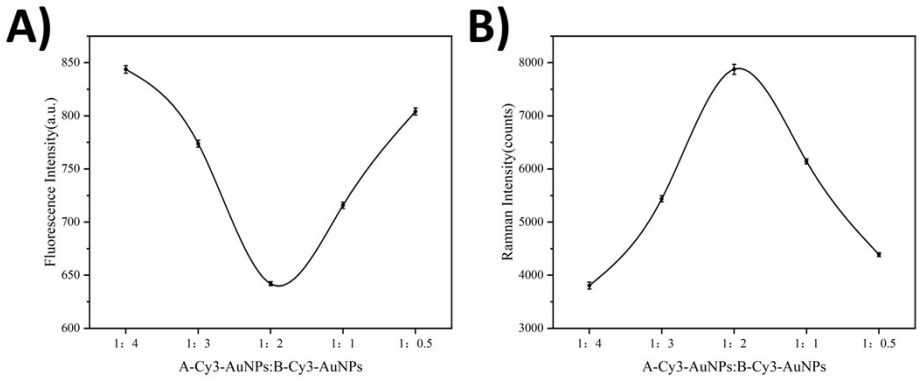

Fig. S4 Effect of the ratio of substrate A to substrate B on the (A) FL intensity and (B) SERS intensity for 100 pM Pb^{2+}.

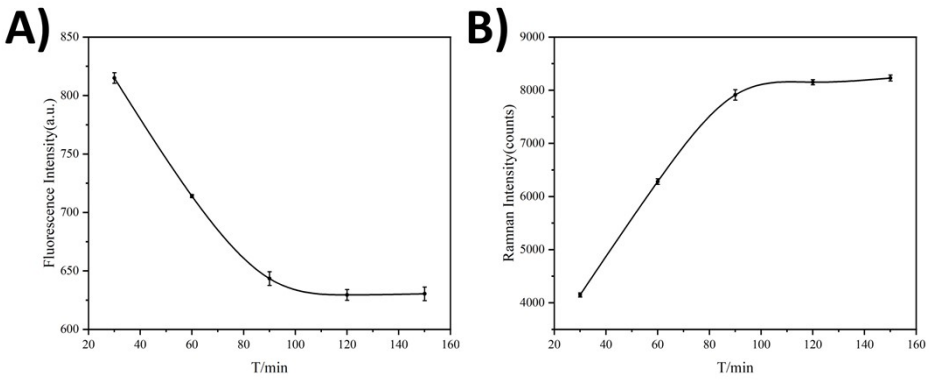

Fig. S5 Effect of the the nonlinear HCR reaction time on the (A) FL intensity and (B) SERS intensity for 100 pM Pb^{2+}.

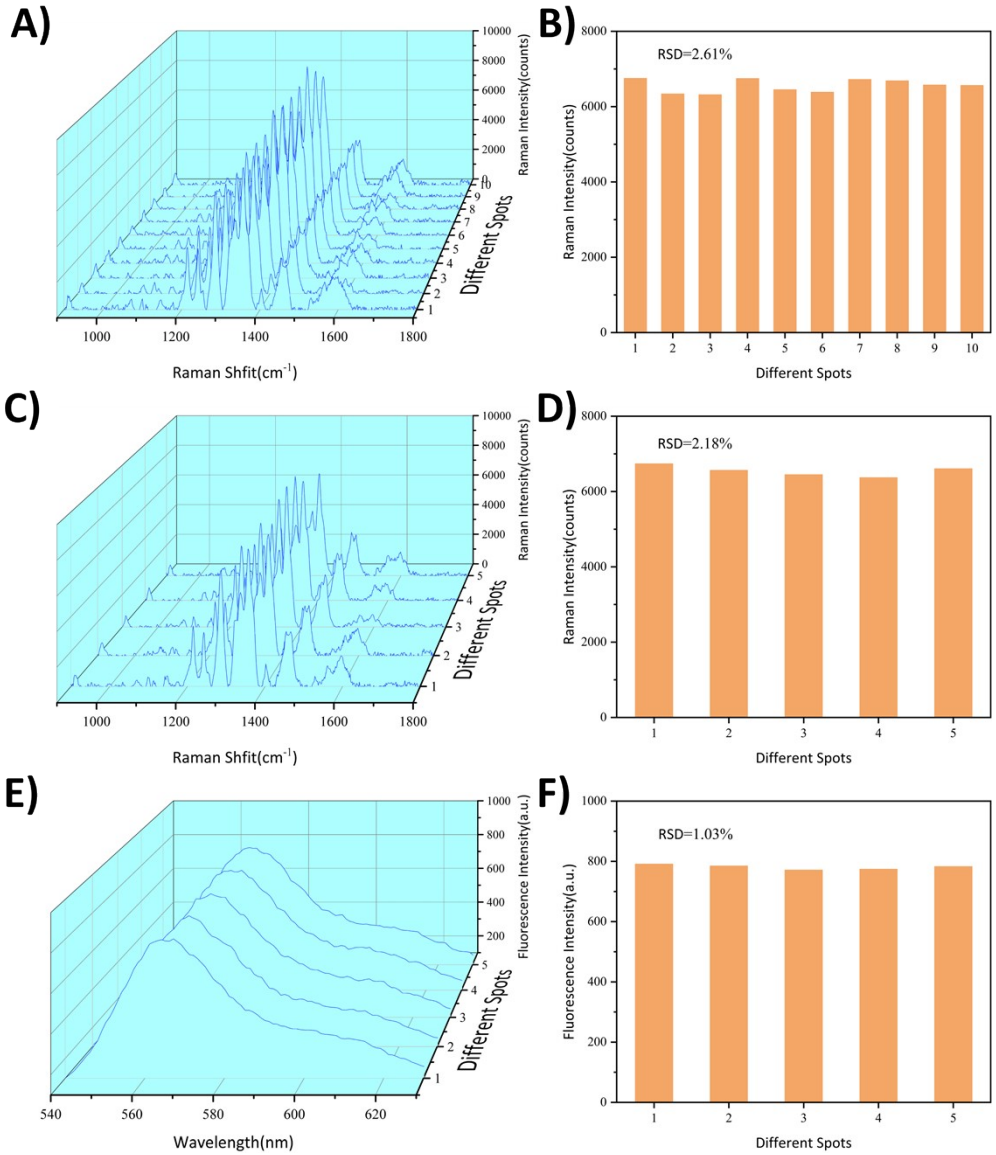

Fig. S6 (A) the Raman spectra of 10 random spots on the SERS mode; (B) RSD of 10 random spots on the SERS mode; (C) the Raman spectra of 5 parallel SERS modes; (D) RSD of 5 parallel SERS modes; (E) the FL spectra of 5 parallel FL modes; (F) RSD of 5 parallel FL modes.