Electronic Supplementary Material (ESI) for Analyst.

Label-Free Technology for the Amplified Detection of MicroRNA Based on the Allosteric Hairpin DNA Switch and Hybridization Chain Reaction

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Table S1. DNA and RNA sequences used in this work.

Name	Sequence
Hairpin DNA	5’-ATTGACCGCTGTGTGACGCAACACTCAATAACTATACAACCTACTACCTCAGTTATTGAGTGTTCGATTCGGCGTG-3’
C1-let7	5’-AGTGTTCGATTCGGCGTGGGTTAACACGCCGAATCGAACACTCAATAAC-3’
C2-let7	5’- TTAACCCACGCCGAATCGAACACTGTTATTGAGTGTTCGATTCGGCGTG-3’
Hairpin DNA-G5	5’-ATTGACCGCTGTGTGACGCAACACTCAATAACTATACAACCTACTACCTCAGTTATTGAGTGTTACTAGGATTCGGCGTG-3’
C1-let7-G5	5’-TTACTAGGATTCGGCGTGGGTTAACACGCCGAATCCTAGTAACACTCA-3’
C2-let7-G5	5’-TTAACCCACGCCGAATCCTAGTAATGAGTGTTACTAGGATTCGGCGTG-3’
Hairpin DNA-G13	5’-ATTGACCGCTGTGTGACGCAACACTCAATAACTATACAACCTACTACCTCAGTTATTGAGTGTTCAAAGTAGTCTAGGATTCGGCGTG-3’
C1-let7-G13	5’-AGTCTAGGATTCGGCGTGGGTTAACACGCCGAATCCTAGACTACTTTG-3’
C2-let7-G13	5’-TTAACCCACGCCGAATCCTAGACTCAAAGTAGTCTAGGATTCGGCGTG-3’
let-7 a	5’-UGAGGUAGUAGGUUGUAUAGUU-3’
let-7 b	5’-UGAGGUAGUAGGUUGUGUGGUU-3’
let-7 c	5’-UGAGGUAGUAGGUUGUAUGGUU-3’
let-7 d	5’-AGAGGUAGUAGGUUGCAUAGU-3’
let-7 e	5’-UGAGGUAGGAGGUUGUAUAGU-3’
let-7 f	5’-UGAGGUAGUAGAUUGUAUAGUU-3’
let-7 g	5’-UGAGGUAGUAGUUUGUACAGU-3’
let-7 i	5’-UGAGGUAGUAGUUUGUGCUGUU-3’
Hairpin DNA-miRNA 21	5’-ATTGACCGCTGTGTGACGCAACACTCAATTCAACATCAGTCTGATAAGCTATGAATTGAGTGTTACTAGGATTCGGCGTG-3’
miRNA 21	5’-UAGCUUAUCAGACUGAUGUUGA-3’

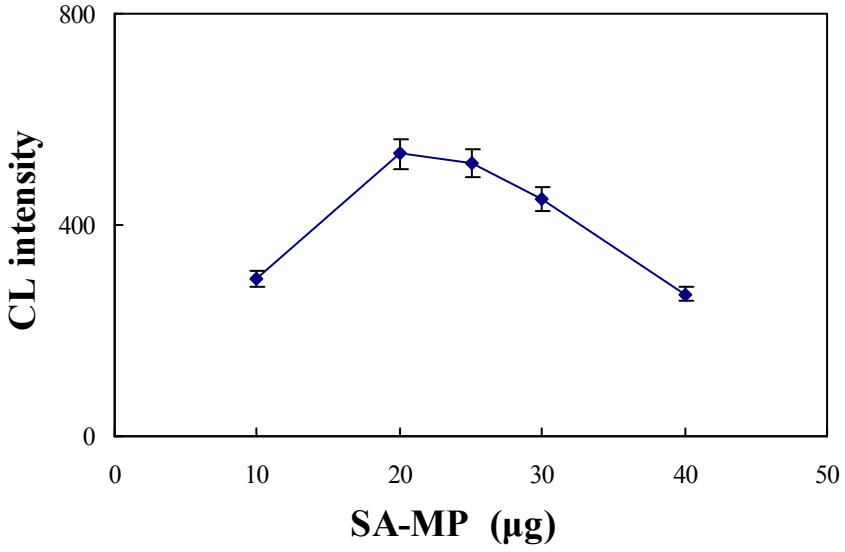


Figure S1. CL intensity vs the amount of SA-MPs. Experimental conditions: aMB probe, C1 and C2 sequences were 10, 20 and 30 pmol. 20 mM Mg^{2+} ions in BA and 100 fmol let 7a, respectively. The detection procedure was carried out as described in the Experimental section

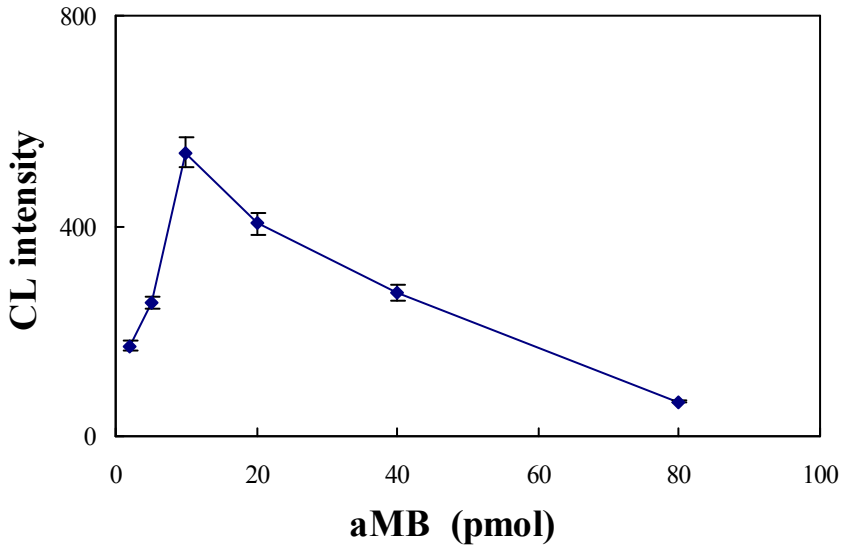


Figure S2. CL intensity vs the amount of aMB probes. Experimental conditions: SA-MPs were 20 µg, aMB probe, C1 and C2 sequences were 10, 20 and 30 pmol. 20 mM Mg^{2+} ions in BA and 100 fmol let 7a, respectively. The detection procedure was carried out as described in the Experimental section.

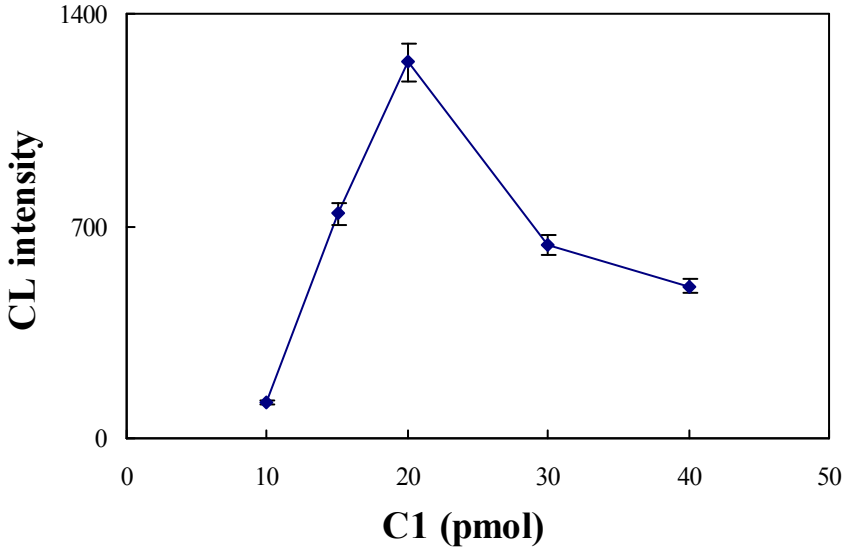


Figure S3. CL intensity vs the amount of C1. Experimental conditions: SA-MPs were 20 μg, aMB probe and C2 sequences were 10 and 20 pmol. 20 mM Mg^{2+} ions in BA and 100 fmol let 7a, respectively. The detection procedure was carried out as described in the Experimental section.

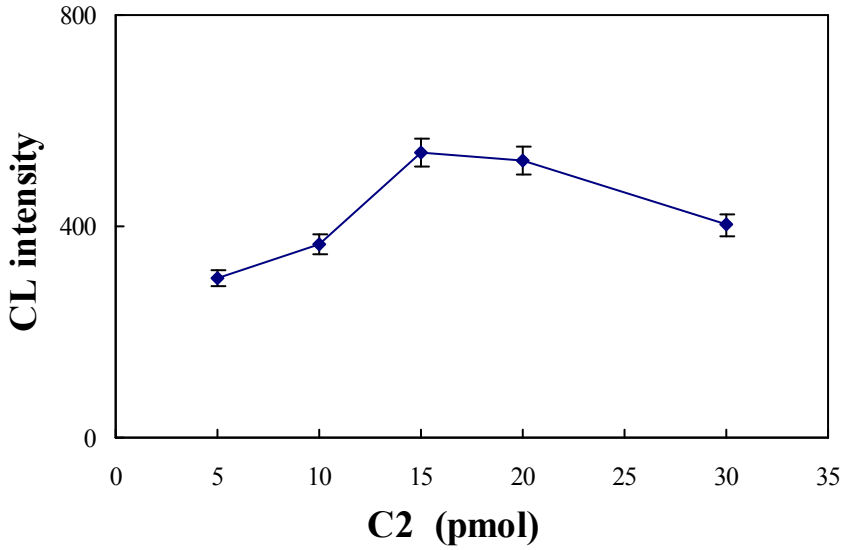


Figure S4. CL intensity vs the amount of C2. Experimental conditions: SA-MPs were 20 μg, aMB probe and C1 sequences were 10 and 20 pmol. 20 mM Mg^{2+} ions in BA and 100 fmol let 7a, respectively. The detection procedure was carried out as described in the Experimental section.

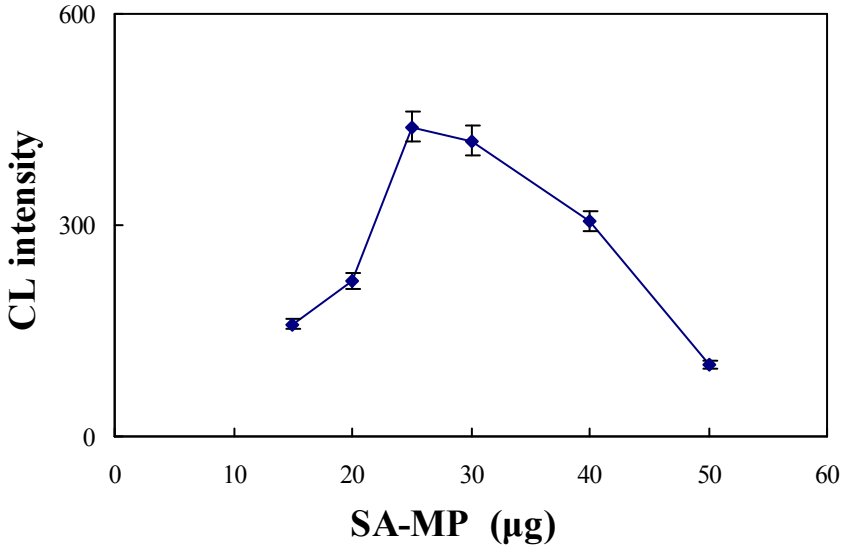


Figure S5. CL intensity vs the amount of SA-MPs. Experimental conditions: aMB probe and let 7a were 10 and 1 pmol, respectively. The detection procedure was carried out as described in the Experimental section.

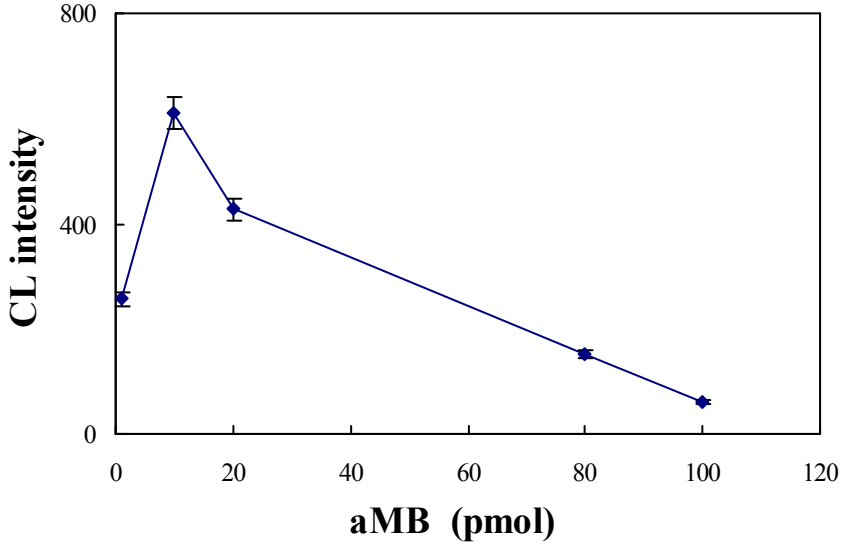


Figure S6. CL intensity vs the amount of aMB probes. Experimental conditions: SA-MPs were 20 μg, let 7a was 1 pmol, respectively. The detection procedure was carried out as described in the Experimental section.

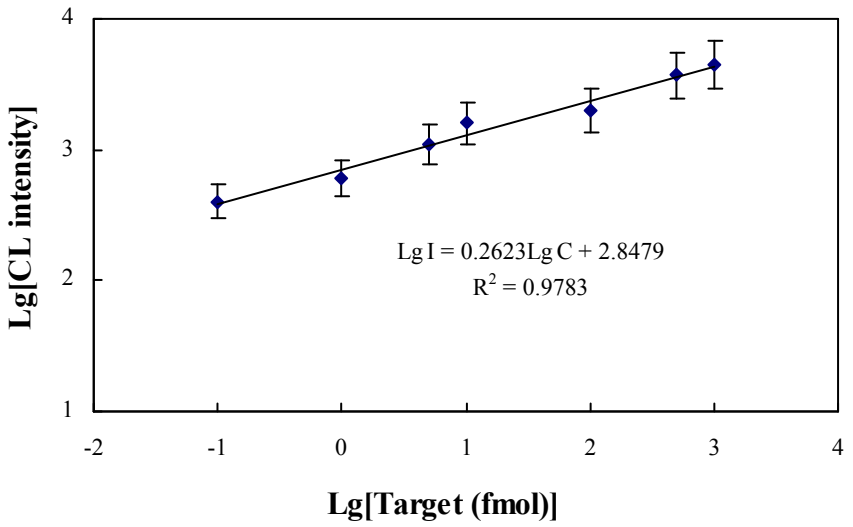
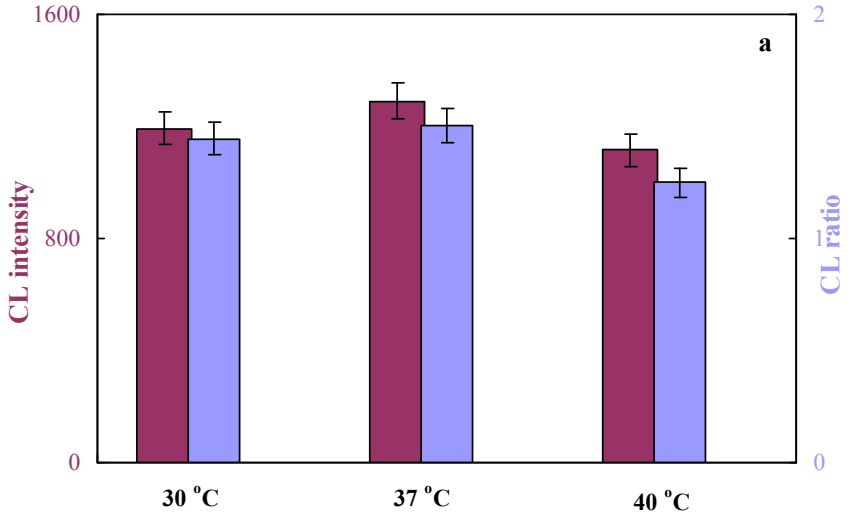


Figure S7. Log-Log calibration data for the target miRNA 21. Experimental conditions: SA-MPs were 20 μg, hairpin DNA-miRNA 21 switch probe, C1 and C2 sequences were 10, 20 and 20 pmol, 20 mM Mg^{2+} ions were in BA, respectively. The detection procedure was carried out as described in the Experimental section.



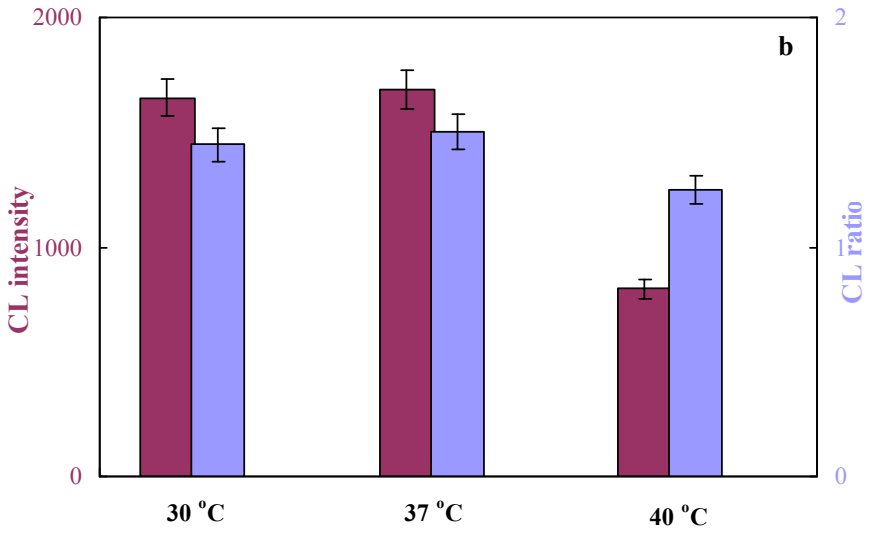


Figure S8. CL intensity and CL ratio vs different temperatures for let 7a (a) and miRNA 21 (b). Experimental conditions: SA-MP was 20 µg, both let-7a and miRNA 21 were 100 fmol, hairpin DNA switch probe, C1 and C2 sequences were 10, 20 and 20 pmol, 20 mM Mg^{2+} ions were in BA, respectively. The detection procedure was carried out as described in the Experimental section.

Table S2. Comparison of sensitivity for different RNA assay methods.

Analytical method	Label	target RNA	Detection Limit
Electrochemical detection	Alkaline Phosphatase (ALP)	p185 BCR-ABL	1 fM [1]
Fluorescence	Poly(dimethylsiloxane)	Dengue virus	50 pM [2]
Fluorescence	SYBR Green I,	Let-7a	10 fM [3]
Surface plasmon resonance (SPR)	Label-free	miR24, 96, 1, 424	10 pM[4]
Fluorescence	Cy3	Let-7a, miR21, 96, 206, 31	10 fM [5]
Silicon photonic microring resonators	Label-free	Let-7a	150 fmol [6]
Polyacrylamide gel electrophoresis/ MS	Label-free	RNase P, 7SL RNA and U6 snRNA,	100 fmol [7]
Capillary electrophoresis	Label-free	Nat-siRNAATGB2	35 amol [8]
Colorimetry / Fluorescence	Methyl / Rhodamine Red	Genomic RNA	1 pmol [9]
Electrochemical detection	HRP	miRNA-21	60 fmol [10]
SPR	Label-free	mRNA from L. Innocua iap	200 pM [11]
SPR imaging	Label-free	16S ribosomal RNA	20 fmol [12]
Nanomechanical detection	Label-free	Messenger RNA of aldolase A	10 pM [13]
CL technique (this work)	Label-free	22 bases RNA	0.1 fmol (1 pM)

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