Electronic Supplementary Material (ESI) for Analyst.

Supporting Information

A paper-supported sandwich immunosensor based on upconversion luminescence resonance energy transfer for the visual and quantitative determination of a cancer biomarker in human serum

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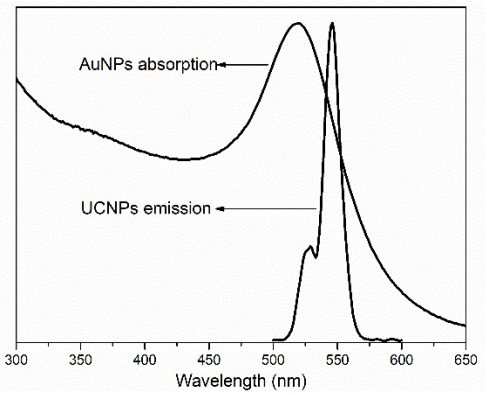

Fig. S1 The fluorescence emission spectrum of UCNPs and the UV-vis absorption spectrum of AuNPs.

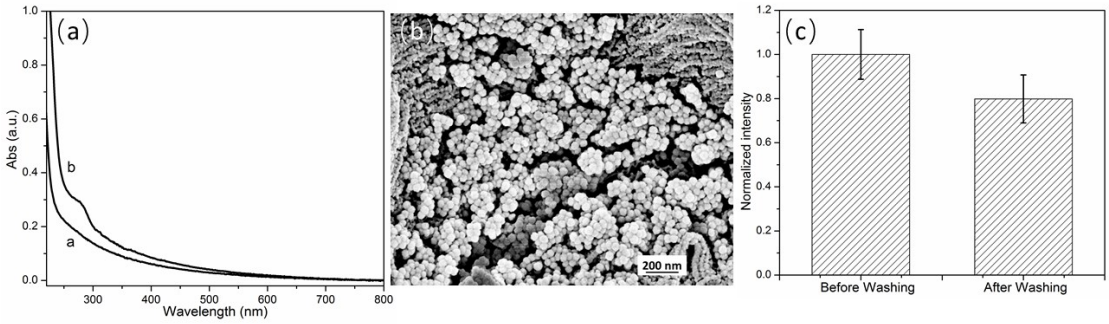

Fig. S2 (a) The UV-vis absorption spectra of UCNPs (curve a) and the UCNPs-CEA Ab 1 (curve b). (b) The SEM image of the as-prepared UCNPs-CEA Ab 1 immobilized on the surface of paper. (c) The normalized intensity of UCNPs-CEA Ab 1 on the surface of paper before and after washing by PBS buffer for several times. The luminescence intensity was normalized to the intensity before washing. Data are presented as the average ± SD from three independent measurements.

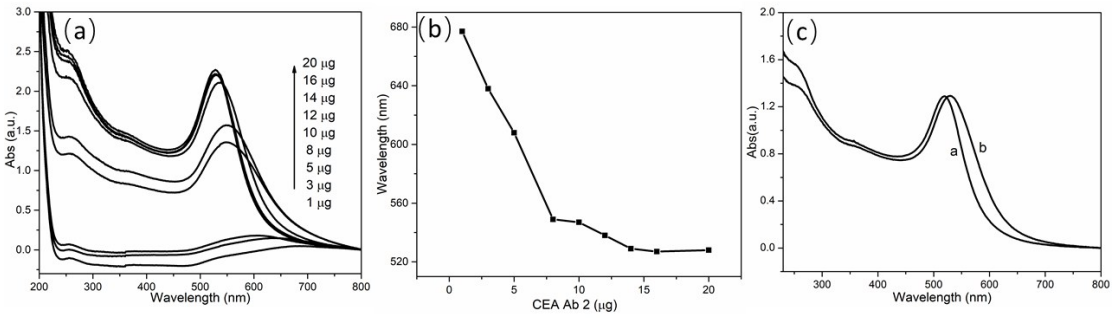

Fig. S3 (a) The UV-vis absorption spectra of AuNPs conjugated with the increasing amounts of CEA Ab 2 (1, 3, 5, 8, 10, 12, 14, 16, 20 μg, respectively). (b) The peak position corresponding to the maximum absorption of AuNPs conjugated with the increasing amounts of CEA Ab 2 (1, 3, 5, 8, 10, 12, 14, 16, 20 μg, respectively). (c) The UV-vis absorption spectra of AuNPs (curve a) and AuNPs-CEA Ab 2 (curve b) conjugated with 16.8 μg CEA Ab 2.

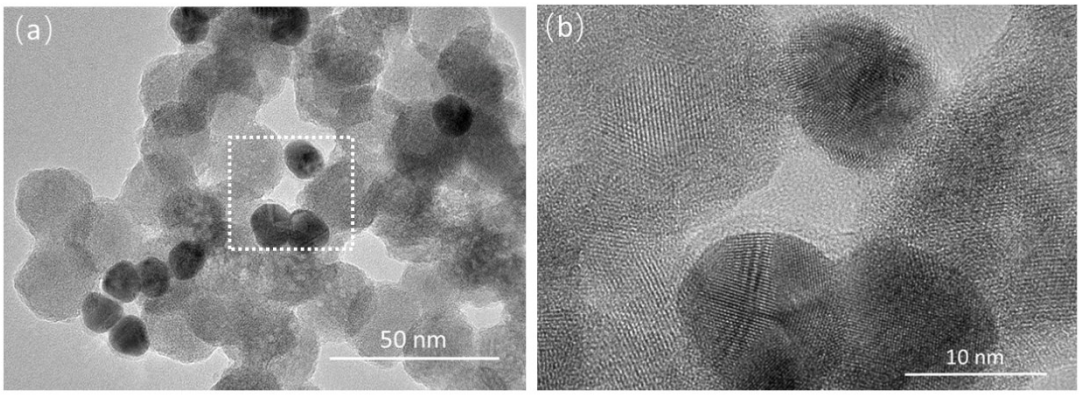

Fig. S4 (a) TEM image of UCNPs-CEA Ab 1-CEA-AuNPs-CEA Ab 2 complex. (b) HRTEM image of UCNPs-CEA Ab 1-CEA-AuNPs-CEA Ab 2 complex.

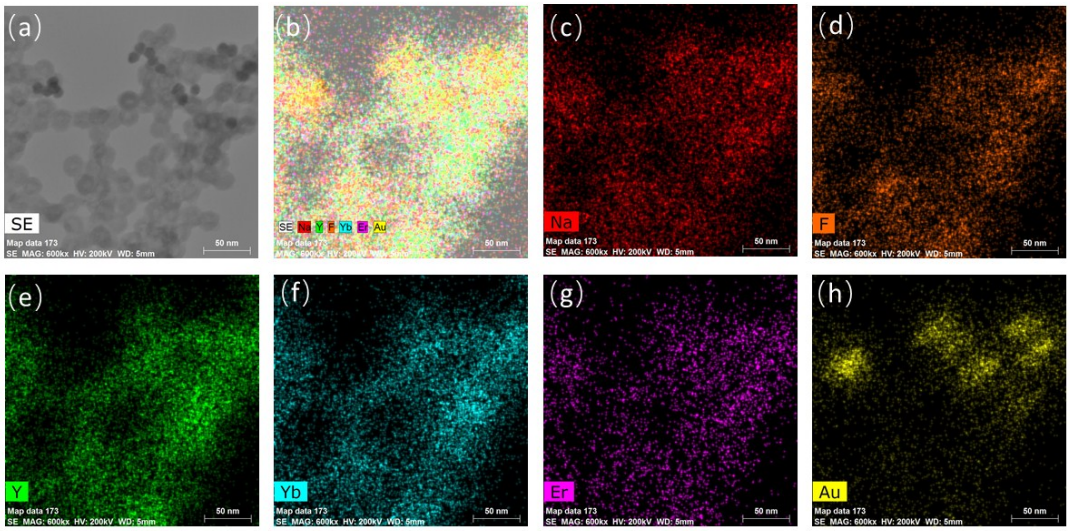

Fig. S5 EDX element mapping analysis of UCNPs-CEA Ab 1-CEA-AuNPs-CEA Ab 2 complex.

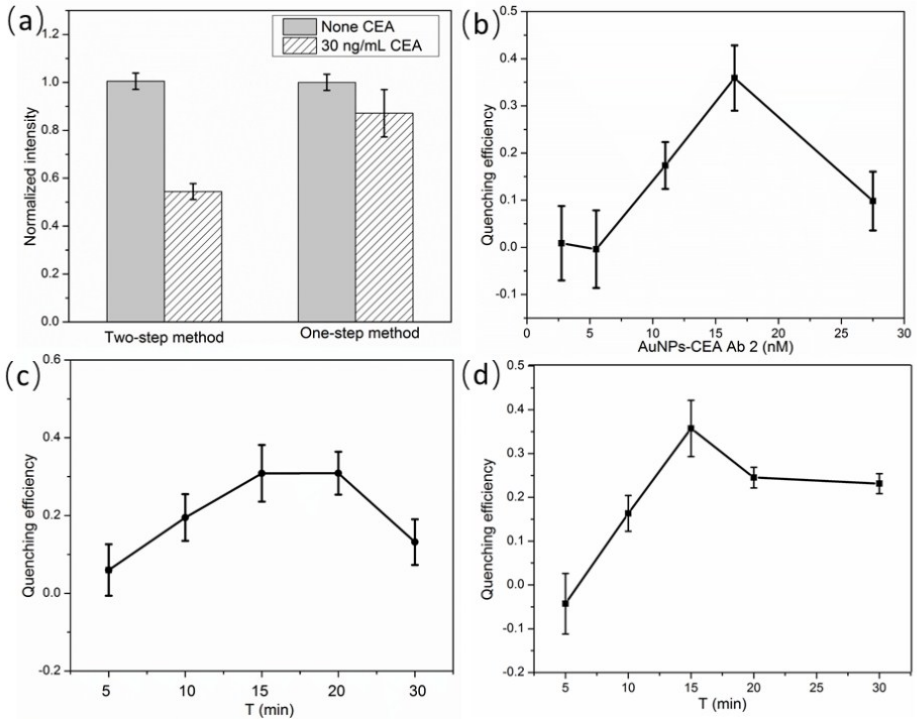

Fig. S6 (a) The normalized intensity in the test zones with 0 and 30 ng/mL CEA and 16.5 nM AuNPs-CEA Ab 2, in which CEA and AuNPs-CEA Ab 2 were added separately and simultaneously. (b) The quenching efficiency in the test zones with 20 ng/mL CEA and different amounts of AuNPs-CEA Ab 2 (0, 5.5, 11, 16.5, 27.5 nM). (c) CEA reaction time dependence of quenching efficiency in the test zones with 20 ng/mL CEA and 16.5 nM AuNPs-CEA Ab 2. (d)

AuNPs-CEA Ab 2 reaction time dependence of quenching efficiency in the test zones with 20 ng/mL CEA and 16.5 nM AuNPs-CEA Ab 2.

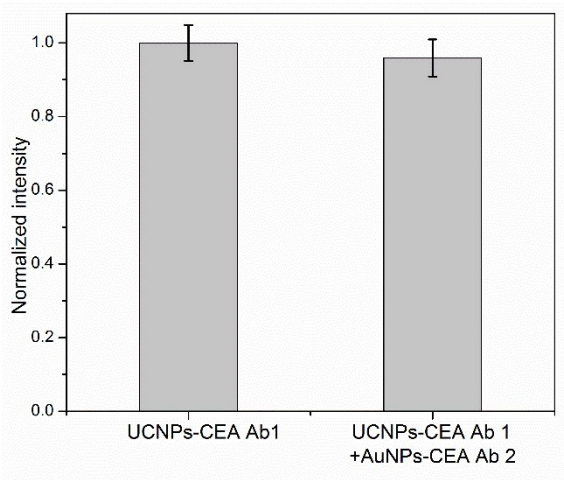

Fig. S7 The normalized intensity of UCNPs-CEA Ab 1 on the surface of paper before and after added 16.5 nM AuNPs-CEA Ab 2, which were washing by PBS buffer for several times. The luminesence intensity was normalized to the intensity without AuNPs-CEA Ab 2. Data are presented as the average ± SD from three independent measurements.

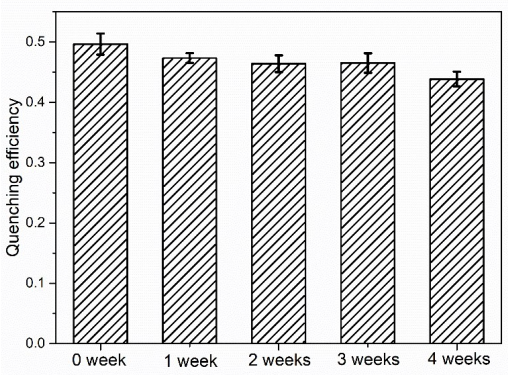

Fig. S8 The quenching efficiency in the test zones with 30 ng/mL CEA and 16.5 nM AuNPs-CEA Ab 2, which were kept in 4℃ dry environment for different time. Data are presented as the average ± SD from three independent measurements.

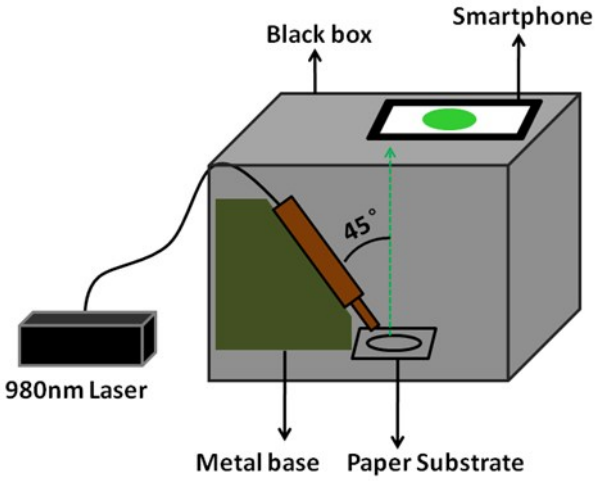

Fig. S9 Illustration of the self-made device for the collection of upconversion luminescence images, which consists of a 980 nm diode laser as the light source, a homemade metal base as sample holder, a black box and a smartphone.

Table S1 A detailed comparison on analytical performance for CEA detection between previously reported methods and this method.

Detection methods	Probes	Linear range (ng/mL)	LOD (ng/mL)	Detection time (h)	Ref.
Fluorescence Immunoassay	UCNPs/ magnetic beads	0.05-20	0.05	＞2.5	[1]
Homogeneous UC-FRET	UCNPs/CNPs	0.1–40	0.1	3	[2]
Homogeneous UC-FRET	UCNPs/AuNPs	0.05-2	0.02	2.5	[3]
Photoelectrochemical Aptasensor	UCNPs@TiO_2 microrods/RCA	0.01-40	0.0036	6	[4]
Photoelectrochemical Aptasensor	UCNPs/Ag_2S/HCR	0.005-5.0	0.0019	2.5	[5]

Paper-based UC-FRET	UCNPs/AuNPs	1-100	0.89	Not mentioned	[6]
FRET-based lateral flow test strip	FITC/AuNPs	5-80	5	0.08	[7]
Paper-based UC-FRET	UCNPs/AuNPs	0.5-30	0.21	＜1	This work

References

[1] Y. H. Li, Z. J. Wu, Z. H. Liu, Analyst, 2015, 140, 4083.

[2] Z. J. Wu, H. Li, Z. H. Liu, Sensors and Actuators B, 2015, 206, 531.

[3] X. Li, W. Sun, J. Wu, Y. Gao, J. Chen, M. Chen, Q. Ou, Anal. Methods, 2018, 10, 1552.

[4] Z. L. Qiu, J. Shu, D. P. Tang, Anal. Chem., 2018, 90, 1021.

[5] Z. L. Qiu, J. Shu, D. P. Tang, Anal. Chem., 2018, 90, 12214.

[6] S. Xu, B. Dong, D. L. Zhou, Z. Yin, S. B. Cui, W. Xu, B. J. Chen, H. W. Song, Sci. Rep., 2016, 23406.

[7] J. D. Wang, F. J. Cao, S. L. He, X. Y. Liu, W. X. Jiang, Y. Y. Yu, H. S. Zhang, W. W. Chen, Talanta, 2018, 176, 444.