Supplementary Materials

SERS-based Dynamical Monitoring of Minimal Residual Disease Markers with a High Sensitivity for Clinical Applications

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SI. Instruments

SERS measurements were performed under a confocal microscope (FV 1000, Olympus). The Raman scattering light was directed to an Andor shamrock spectrograph equipped with a charge-coupled device (CCD). He–Ne laser (Melles Griot, 05-LHP-991) with 632.8 nm radiation was used for excitation and the laser power at the sample position was 2.38 mW. All the spectra here were the results of an accumulation of 60 s. Transmission electron microscope (TEM) images and energy-dispersive X-ray (EDX) spectrum were obtained with an FEI Tecnai G2T20 electron microscope operating at 200 kV. A high speed centrifuge (2-16 PK, Sigma, Germany) was used for the purification of samples.

Supporting Figures

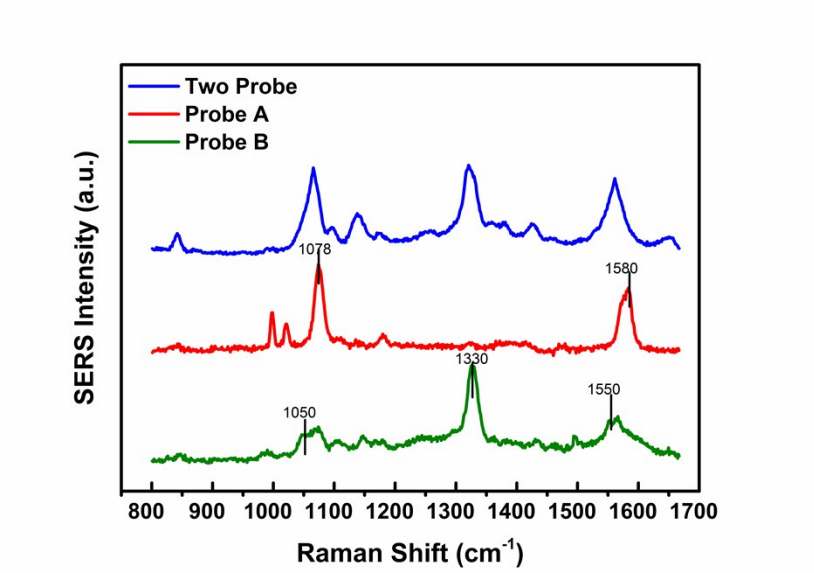

Fig. S1 Representative SERS spectra of Probe A (red), Probe B (green) and their equivalent mixture (blue).

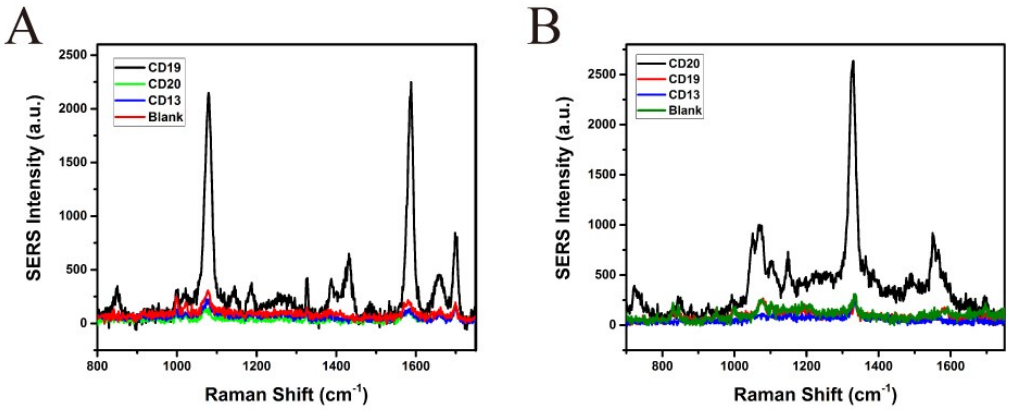

Fig. S2 SERS spectra of Probe A (A) and Probe B (B) after incubated with CD19, CD20, CD13 proteins and PBS solution.

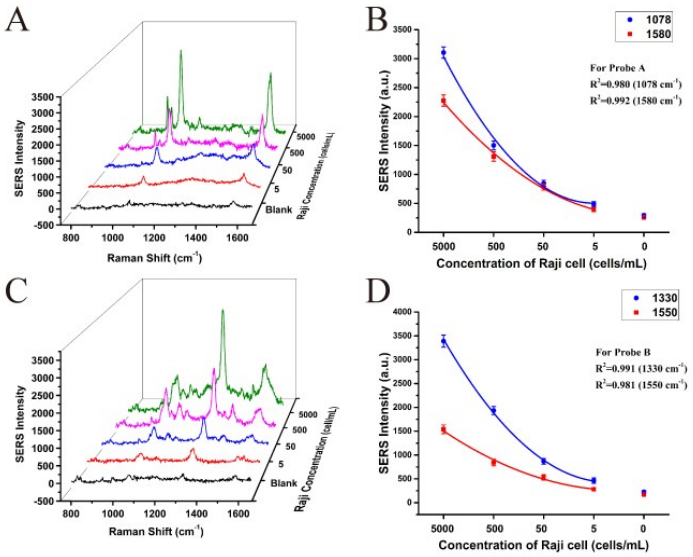

Fig. S3 SERS spectra for the separate detection of CD 19 (A) and CD20 (C) in Raji cells at different concentrations, and the calibration curves at 1580 and 1078 cm-1 (B), 1330 and 1550 cm-1 (D). The error bars indicate the standard deviations of five replicated measurements.

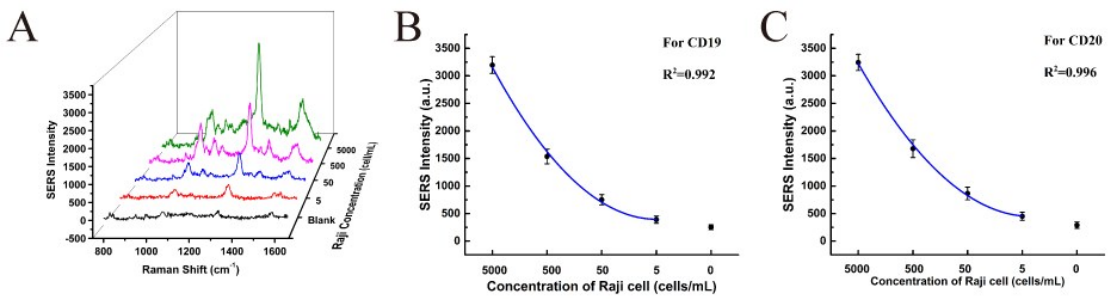

Fig. S4 (A) The simultaneous detection of the two markers in Raji cells of different concentrations. Concentration-related SERS signals of peak intensity at 1078 cm^{-1} for CD19 (B) and that at 1330 cm^{-1} for CD20 (C) in the simultaneous detection. The error bars indicate standard deviations of five replicated measurements. The regression equations are as follows: $y=5297.210-2446.042x+305.220x^2$, $R^2=0.992$ (for CD19); $y=5247.679-2300.131x+275.544x^2$, $R^2=0.996$ (for CD20); x axis, concentrations of Raji cell; y axis, SERS intensity.

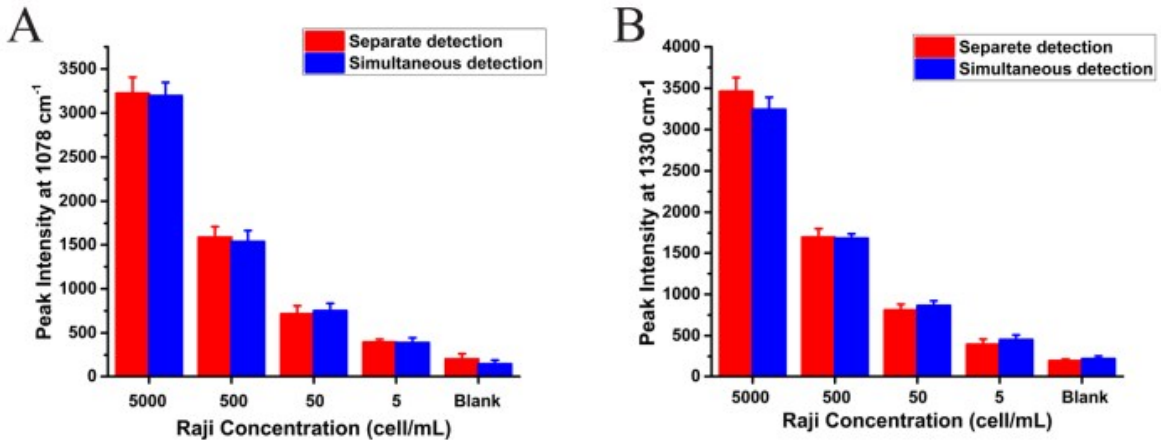

Fig. S5 Comparisons of the SERS intensity of separate (blue) and simultaneous (red) detections for CD19 (A) and CD20 (B). Two types of SERS probes with a volume ratio of 1: 1 were mixed together for simultaneous detection. The error bars represent the standard deviation of five measurements.

Supporting Table:

NO.	Sex	Age	diagnosis	FCM (CD19) Before/After	FCM (CD20) Before/After
1	Female	10	B-leukemia	1.5% / 0%	2.6% / 0%
2	Male	54	B-lymphoma	0.6% / 0%	0.8% / 0%
3	Female	33	B-leukemia	0.7% / 0%	0.4% / 0%
4	Male	38	B-lymphoma	18.8% / 0.9%	6.3% / 1.3%
5	Male	76	B-lymphoma	20.1% / 2.5%	12.3% / 2.2%
6	Female	55	B-leukemia	2.2% / 1.4%	0.2% / 0.4%
7	Female	40	B-leukemia	2.9% / 2.6%	2.5% / 5.4%
8	Male	12	B-lymphoma	3.2% / 3.1%	19.9% / 3.6%
9	Male	7	B-leukemia	4.8% / 7.2%	8.2% / 5%
10	Female	61	B-lymphoma	6.9% / 19.1%	0.4% / 14.3%
11	Male	68	B-lymphoma	11% / 17.8%	10.8% / 8.8%
12	Female	42	B-lymphoma	10.3% / 10.4%	2.1% / 17.6%
13	Male	25	B-lymphoma	9.8% / 9.3%	0.1% / 9.2%

Table S1 The demographics and diagnosis characteristics of 13 clinical samples.