

Supporting Information for

**Fork-shaped Paper SERS Sensor Coated with Raspberry-like
Bimetal Nanospheres for Boosted Mixture Detection:
Experimental Design and Application**

Yuanyuan Xu¹, Xingguo Gao¹, Cheng Yang², Baoyuan Man^{2*}, Jiancai
Leng^{1*}

1. School of Science, Qilu University of Technology (Shandong Academy of Sciences), Jinan 250353, People's Republic of China.
2. School of Physics and Electronics, Shandong Normal University, Jinan 250014, People's Republic of China.

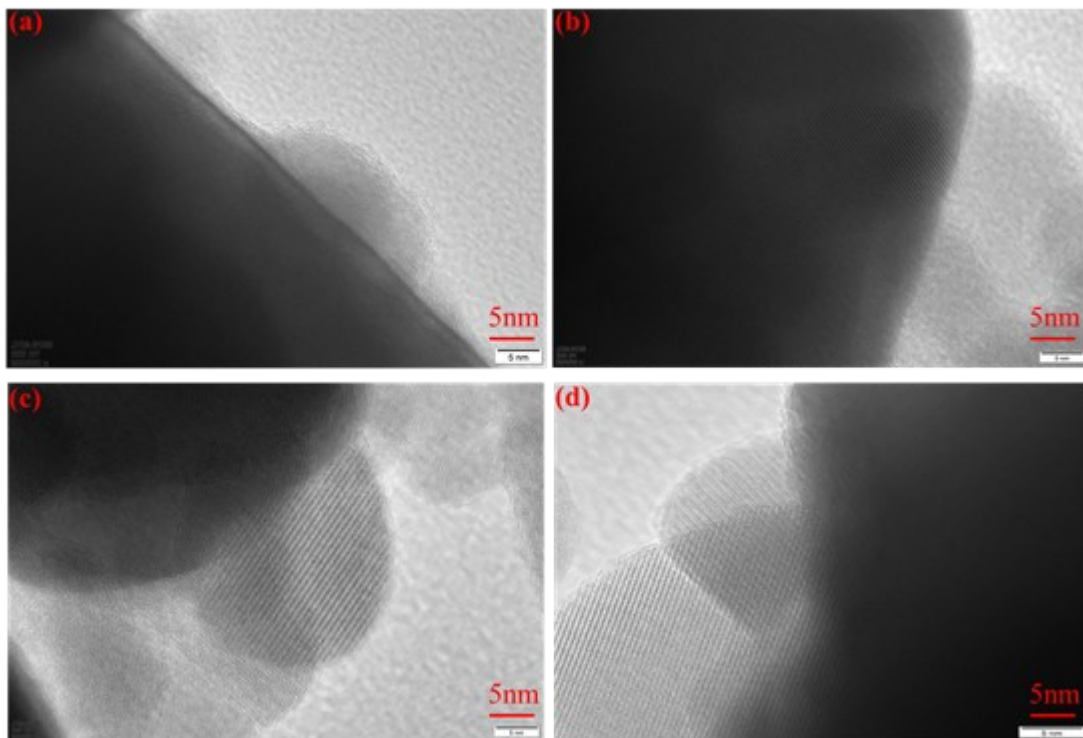


Fig. S1. High resolution TEM images of raspberry-like bimetal Au@AgNPs.

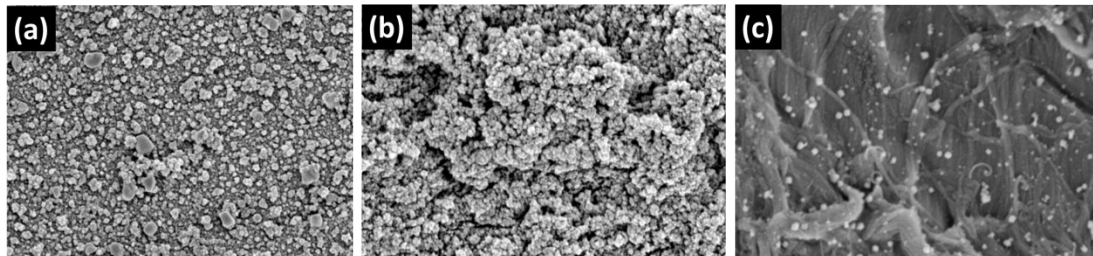


Fig. S2. SEM images of AgNPs/paper substrates obtained by growth solution of different concentration (a) 0.04M and (c) 0.01M. (b) SEM images of AgNPs/paper (Fig. S2a) after exposing to the HAuCl₄ solution.

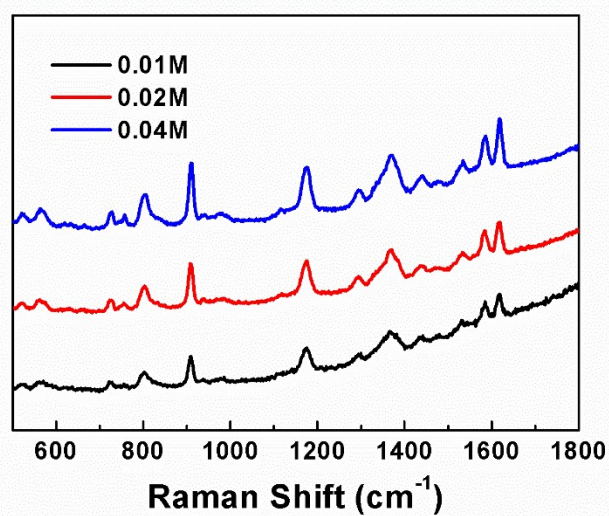


Fig. S3. The Raman spectra of 10^{-7} M CV on different AgNPs/paper substrates.

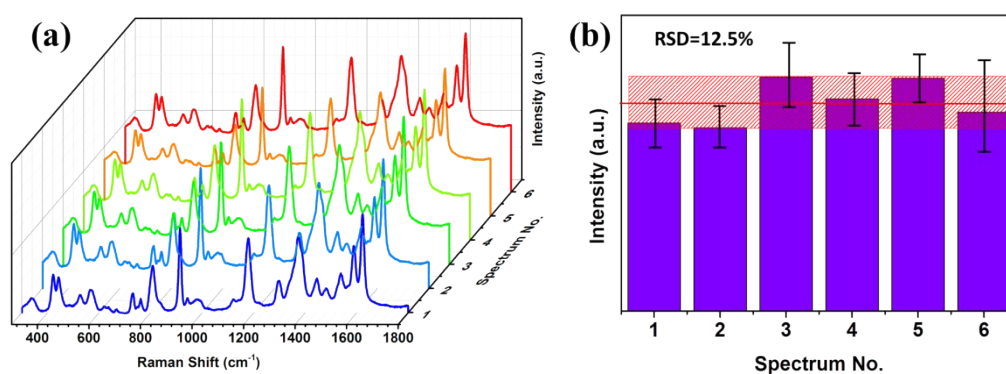


Fig. S4. (a) SERS signals of CV molecules at 10^{-8} M from 6 different batches paper substrates. (b) Intensity distribution of the peak at 914 cm^{-1} .

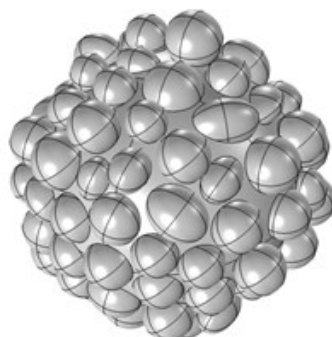


Fig. S5. Model of Raspberry-like bimetal Au@AgNP used in the COMSOL software according to its SEM images.

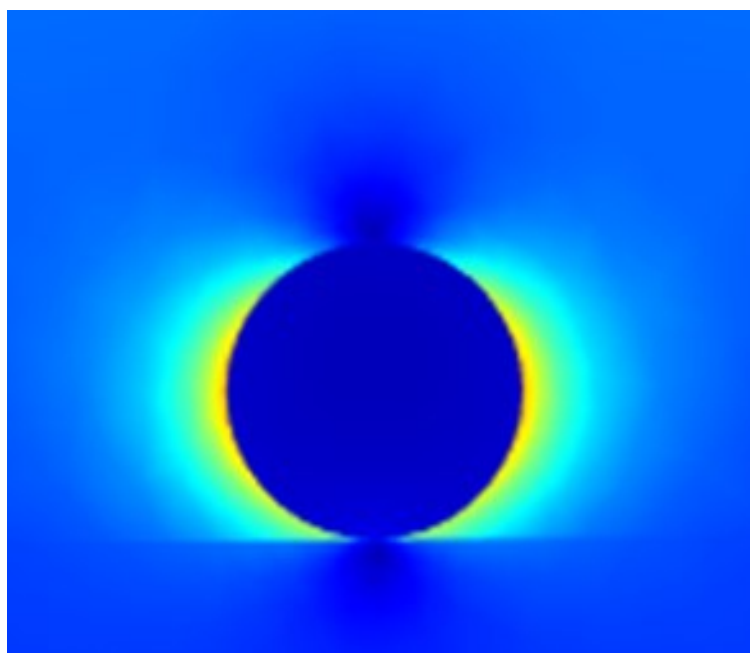


Fig. S6. The electric field distribution on the AgNP.

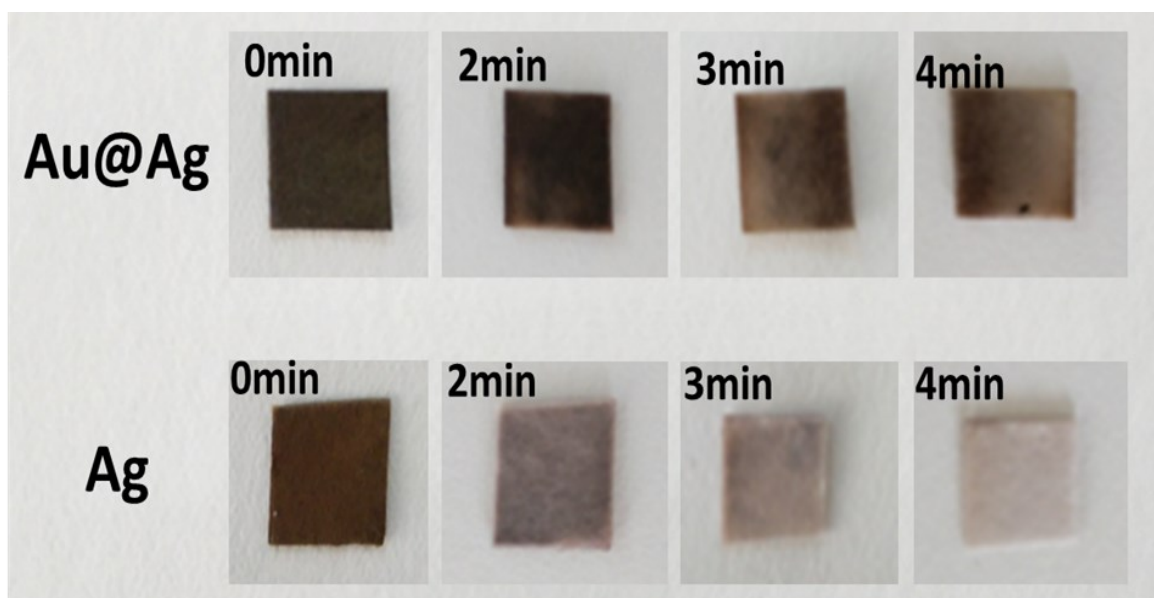


Fig. S7. Optical photos of both paper substrates after immersing in aqueous H₂O₂ for different time.

Table S1. The limit of detections for different mixtures reached in the SERS paper substrates combined paper chromatography.

Substrates	Analytes	LOD	Reference
Ag@SiO ₂ paper	R6G and malachite green	0.5 mM and 0.5 mM	Ref. 1
Ag nanoislands/paper	CR, TB and CV	50 mM, 40 mM and 5 mM	Ref. 2
AuNP/MoS ₂ /filter paper	CV and TB	10 ⁻⁸ M and 10 ⁻⁸ M	Ref. 3
AgNPs/paper	R6G, methylene blue and malachite green	1 mM, 1 mM and 1 mM	Ref. 4
AgNPs /filter paper	R6G, CR, CV and MO	4×10 ⁻⁴ M, 4×10 ⁻⁴ M, 4×10 ⁻⁴ M and 4×10 ⁻⁴ M	Ref. 5
ZnO/AgNPs/paper	Dimethoate and thiuram	54.57 μg/L and 19.16 μg/L	Ref. 6
Raspberry-like bimetal Au@AgNPs/paper	CV and fluorescein	10 ⁻⁹ M and 10 ⁻⁹ M	This work

References:

1. K. Zhang, J. Qing, H. Gao, J. Ji, B. Liu, Coupling shell-isolated nanoparticle enhanced Raman spectroscopy with paper chromatography for multi-components on-site analysis, *Talanta*, 2017, 162, 52-56.
2. H. Jung, M. Park, M. Kang, K.-H. Jeong, Silver nanoislands on cellulose fibers for chromatographic separation and ultrasensitive detection of small molecules, *Light: Science & Applications*, 2016, 5, e16009.
3. Y. Zhao, X. Pan, L. Zhang, Y. Xu, C. Li, J. Wang, J. Ou, X. Xiu, B. Man, C. Yang, Dense AuNP/MoS₂ hybrid fabrication on fiber membranes for molecule separation and SERS detection, *RSC Advances*, 2017, 7, 36516-36524.
4. W.W. Yu, I.M. White, Chromatographic separation and detection of target analytes from complex samples using inkjet printed SERS substrates, *Analyst*, 2013, 138, 3679-3686.
5. J.D. Weatherston, R.K.O. Seguban, D. Hunt, H.-J. Wu, Low-Cost and Simple Fabrication of Nanoplasmonic Paper for Coupled Chromatography Separation and Surface Enhanced Raman Detection, *ACS Sensors*, 2018, 3, 852-857.

6. X. Jin, P. Guo, P. Guan, S. Wang, Y. Lei, G. Wang, The fabrication of paper separation channel based SERS substrate and its recyclable separation and detection of pesticides, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 2020, 240, 118561.