Supporting Information

Sonochemical synthesis of highly-branched flower-like Fe_3O_4@SiO_2@Ag microcomposites and their application as versatile SERS substrates

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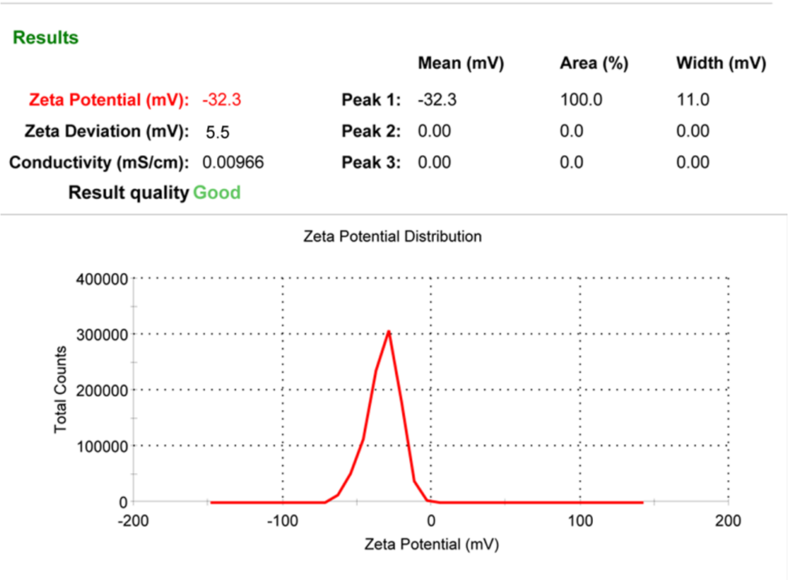

Results

			Mean (mV)	Area (%)	Width (mV)
Zeta Potential (mV):	-32.3	**Peak 1:**	-32.3	100.0	11.0
Zeta Deviation (mV):	5.5	**Peak 2:**	0.00	0.0	0.00
Conductivity (mS/cm):	0.00966	**Peak 3:**	0.00	0.0	0.00
Result quality	Good				

Fig. S1 Zeta potential of Fe_3O_4@SiO_2 microspheres in ethanol.

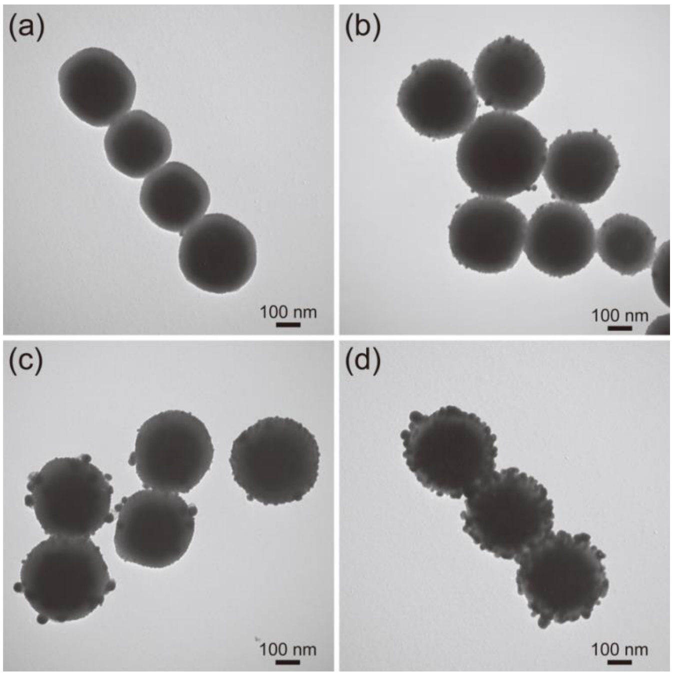

Fig. S2 TEM images of Fe_3O_4@SiO_2–Ag seed synthesized with different concentrations of $AgNO_3$: (a) 0.25, (b) 0.5, (c) 0.75, and (d) 1 mM.

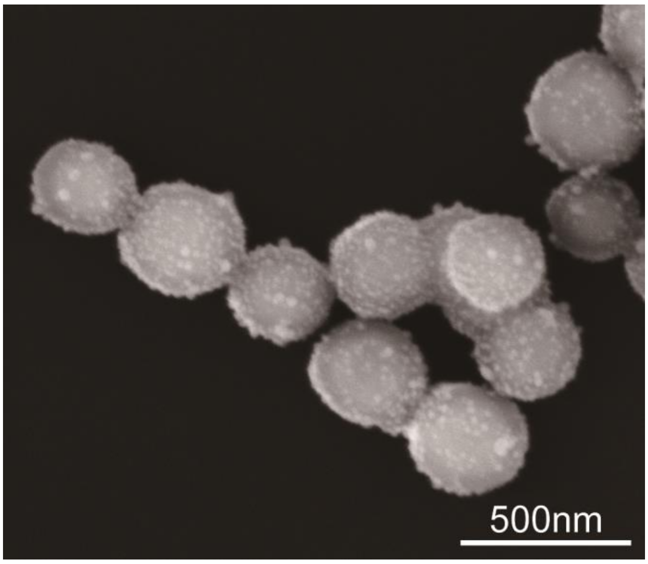

Fig. S3 SEM images of Fe_3O_4@SiO_2–Ag seed particles synthesized with 0.25 mM $AgNO_3$ and butylamine.

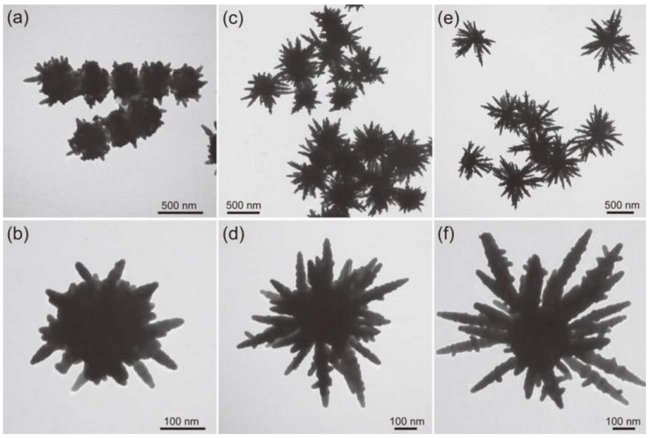

Fig. S4 TEM images of the $Fe_3O_4@SiO_2@Ag$ microflowers synthesized with different concentrations of $AgNO_3$: (a) 0.1 mM, (c) 0.2 mM, and (e) 0.3 mM $AgNO_3$, and their magnified TEM images in (b), (d), and (f), respectively.

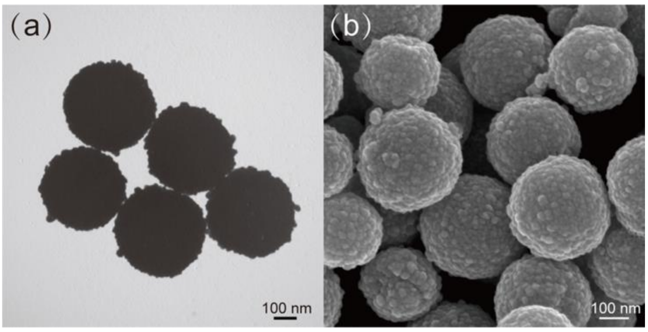

Fig. S5 (a) TEM and (b) SEM images of $Fe_3O_4@SiO_2@Ag$ microspheres synthesized when PVP was added before the addition of CH_2O and $NH_3 \cdot H_2O$.

S6 EF calculation:

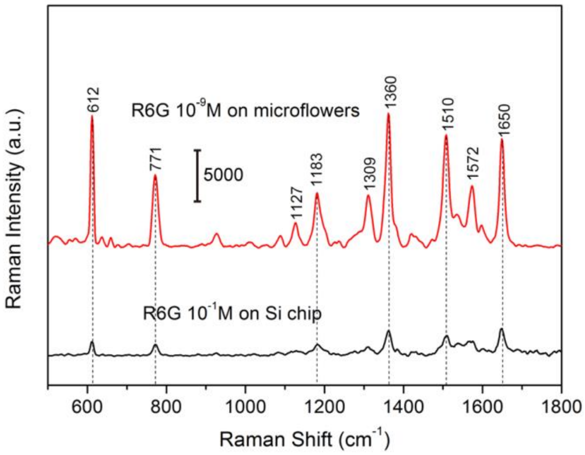

Fig. S6 Raman spectra recorded of R6G molecules with (red) and without (black) $Fe_3O_4@SiO_2@Ag$ microflowers.

To quantify the enhancement ability of proposed microflowers, the enhancement factor (EF) was calculated as the ratio of photons scattered by the SERS substrate and the normal substrate. EF was estimated according to the following equation:

A reliable calculation of the EF through the general formula EF = (I_{SERS}/N_{Surf}) / (I_{RS}/N_{vol}), where N_{Surf} is the average number of R6G molecules contributing to the SERS signal, N_{Vol} is the average number of R6G molecules contributing to the normal Raman signal, and I_{SERS} and I_{RS} are the intensities of the scattering band of interest in the SERS and normal Raman spectra, respectively. However, intrinsic EF is difficult to estimate because several variables, such as adsorbed molecules and laser scattering volume, are difficult to obtain. In our experiment, all the other parameters, including the laser diameter, laser power, exposure time, and microscopic magnification, were identical. The chemical droplets were of the same volume, and the number of detected R6G molecules was proportional to its concentration. Therefore, the EF was roughly estimated by comparing the intensity of the Raman peak in the SERS spectrum with that in the normal Raman spectrum according to the equation EF = $(I_{SERS}/I_{RS}) \times (C_{RS}/C_{SERS})$, where I_{SERS} and I_{RS} are the vibration intensities in the SERS and normal Raman spectra of R6G molecules, and C_{RS} and C_{SERS} are the concentrations of the R6G molecules in the SERS and reference samples, respectively. The peak at 1360 cm^{-1} from the R6G Raman spectrum (Fig. S5) was chosen for analysis, and the intensities for peaks (a) and (d) were 2460 and 12955 a.u., respectively. The R6G concentrations for peaks (a) and (d) were 10^{-1} M and 10^{-9} M, respectively. Therefore, the EF of the Fe_3O_4@SiO2@Ag microflowes was roughly estimated to be 5.26×10^{8}.

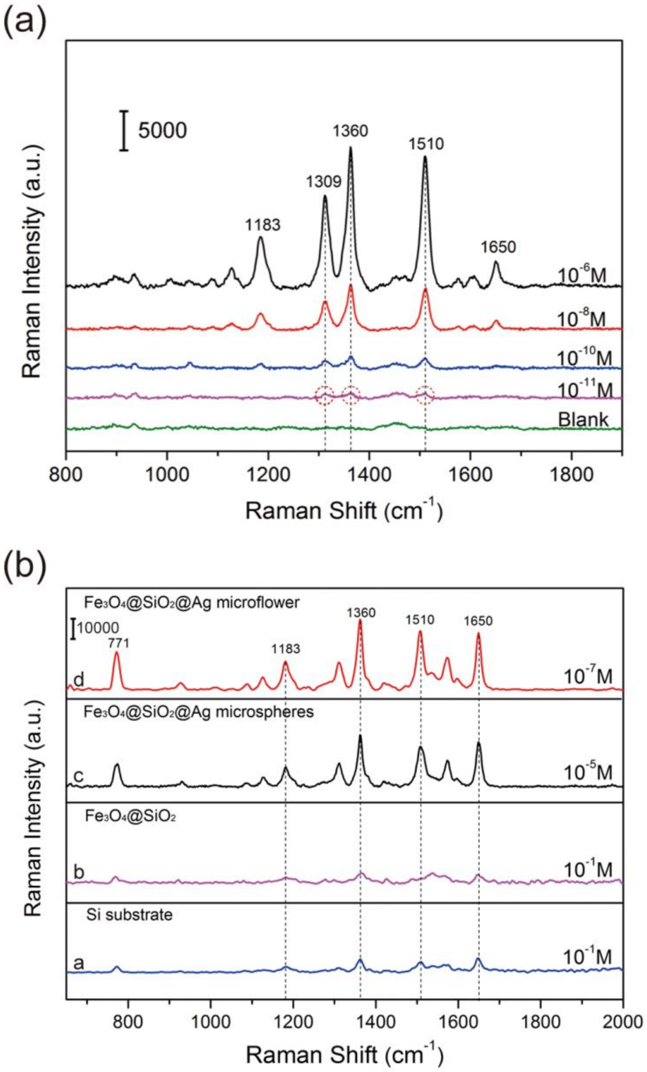

Fig. S7 (a) SERS spectra of R6G at different concentrations on the control Fe_3O_4@SiO_2@Ag microspheres. (b) SERS spectra of R6G on different SERS substrates. a: 10^{-1}M R6G on Si substrate. b: 10^{-1}M R6G on Fe_3O_4@SiO_2 microspheres. c: 10^{-5}M R6G on the control Fe_3O_4@SiO_2@Ag microspheres. d: 10^{-7}M R6G on the Fe_3O_4@SiO_2@Ag microflowers.

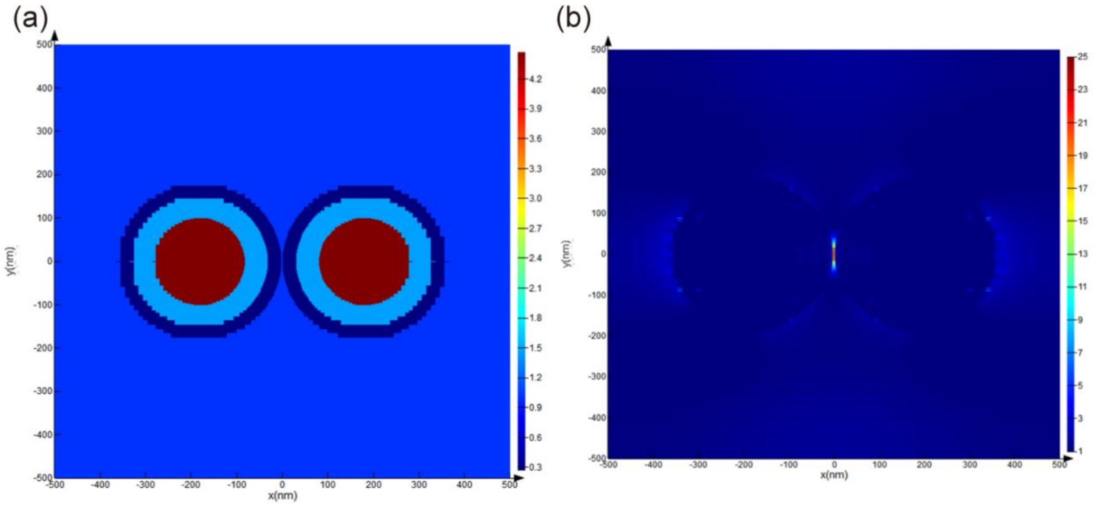

Fig. S8 (a) Simulation model and (b) EM field distributions of the control Fe_3O_4@SiO_2@Ag microspheres.

Table S1

Raman shift (cm^{-1})	Assignment
561	**υs(CSS)**
928	**υ(CH_3N) or υ(C−S)**
1145	**ρ(CH_3) or υ(C−N)**
1385	**ρ(CH_3)**
1515	**υ(C−N)**

Table. S1 Raman peaks of thiram and corresponding assignments.