Supplementary Information

Gap-enhanced Resonance Raman Tags for Live-Cell Imaging

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Figure S1. TEM images of Au cores, P-GERTs and GERRTs. Scale bars are 50 nm for Au cores and P-GERTs, and 100 nm for GERRTs.



Figure S2. SERS spectrum of P-GERTs excited with 785 nm laser, 22 mW power, 2 s acquisition time and $10 \times$ objective lens.



Figure S3. Representative SERS spectra of GERRTs at the concentrations of 1 fM and 0.1 fM.



Figure S4. The intensity of Raman band at 1206 cm⁻¹ of 100 measurements from 10 fM (left), 1 fM (middle), and 0.1 fM (right) GERRTs solution. Red dotted lines represent the background Raman signal (1206 cm⁻¹) without GERRTs.



Figure S5. SERS spectra of the other six single GERRTs measured using the RISE system. The right is the same with the left except the exclusion of the Raman band at 523 cm⁻¹ to zoom in on other bands.

Raman tag	Zeta potential (mV)	Average (mV)
GERRTs	60.2	59.0 ± 2.5
	60.8	
	55.9	
GERRTs@PDA	54.3	50.8 ± 2.5
	49.3	
	48.7	
GERRTs@PDA@PEG	7.9	10.1 ± 1.7
	10.3	
	12.1	

Table S1 | Zeta potentials of GERRTs before and after surface functionalization.