Surface-Enhanced Raman Scattering Based-Molecular Encoding with Gold Nanostars for Anticounterfeiting application

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Supporting Information

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Figure S1 SEM micrograph of Au NSs at 25 kX.



Figure S2 (a) 50 kX SEM image of Au NPs and (b) UV-Vis spectra of Au NP and Au NS solutions.



Figure S3 (a) Raman spectra of Au NSs, Au NSs with 0.5 nM R6G, Au NPs, Au NPs with 0.5 nM R6G. Each sample was measured five times. The excitation wavelength was 1064 nm, laser power was 100%, and the exposure time was 1 second. (b) PCA score plot of the SERS spectra in (a), where each dot stands for an individual spectrum and the ellipses stands for the 95% confidence of where the spectra of this sample should be located.



Figure S4 A local view of PCA score plot, which results from the Raman spectra of eight tags, "unknown" sample, and Au NSs. The "unknown" sample was recognized to be Tag 344 based on the location of 95% confidence ellipse.



Figure S5 Raman spectra of tags with 0-15 nmol 4-MBA and 0-3 nmol MG



Figure S6 Raman spectra of PEO and PEO Au NS films for 1 day, 2 weeks, and 1 year.



Figure S7 Normalized Raman spectra of written tags

| | 4-MBA | R6G | МВ | cv | Melamine | 4-ATP | 4-MPy | Ethanol | MG |
|---------|-------|-----|----|----|----------|-------|-------|---------|----|
| Tag 279 | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 1 |
| Tag 184 | 0 | 1 | 0 | 1 | 1 | 1 | 0 | 0 | 0 |
| Tag 193 | 0 | 1 | 1 | 0 | 0 | 0 | 0 | 0 | 1 |
| Tag 044 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 0 | 0 |
| Tag 006 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 1 | 0 |
| Tag 344 | 1 | 0 | 1 | 0 | 1 | 1 | 0 | 0 | 0 |
| Tag 479 | 1 | 1 | 1 | 0 | 1 | 1 | 1 | 1 | 1 |
| Tag 146 | 0 | 1 | 0 | 0 | 1 | 0 | 0 | 1 | 0 |

Table S1 Coding of tags in Fig.2, where "1" means existence of 1 mM substance and "0" means absence of substance. The "1" and "0" of each tag construct binary codes which can be represented by decimal number shown at left column.

| | 4-MBA | R6G | MB | cv | Melamine | 4-ATP | 4-MPy | Ethanol | MG |
|---------|-------|-----|----|----|----------|-------|-------|---------|----|
| Tag 330 | 1 | 0 | 1 | 0 | 0 | 1 | 0 | 1 | 0 |
| Tag 159 | 0 | 1 | 0 | 0 | 1 | 1 | 1 | 1 | 1 |
| Tag 225 | 0 | 1 | 1 | 1 | 0 | 0 | 0 | 0 | 1 |
| Tag 278 | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 0 |
| Tag 430 | 1 | 1 | 0 | 1 | 0 | 1 | 1 | 1 | 0 |

Table S2 Coding of tags for the four sets of SERS tags in Fig. 3.

| | 4-MBA | R6G | МВ | cv | Melamine | 4-ATP | 4-MPy | Ethanol | MG |
|---------------|-------|-----|----|----|----------|-------|-------|---------|------|
| Tag 279- A | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 1 |
| Tag 279- B | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 0.5 |
| Tag 279- C | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 0.25 |
| Tag 279- D | 1 | 0 | 0 | 0 | 1 | 0 | 1 | 1 | 0 |

Table S3 Coding of tags in Fig. 4, where the number "X" (0, 0.25, 0.5, or 1) means existence of XmM of substance.

| | 4-MBA | R6G | МВ | cv | Melamine | 4-ATP | 4-MPy | Ethanol | MG |
|---------|-------|-----|----|----|----------|-------|-------|---------|----|
| Tag 295 | 1 | 0 | 0 | 1 | 0 | 0 | 1 | 1 | 1 |
| Tag 096 | 0 | 0 | 1 | 1 | 0 | 0 | 0 | 0 | 0 |
| Tag 118 | 0 | 0 | 1 | 1 | 1 | 0 | 1 | 1 | 0 |

Table S4 Coding of tags in Fig. 6 for SERS writable inks.