



## Supplementary Materials: Pt-Based Nanostructures for Observing Genuine SERS Spectra of *p*-Aminothiophenol (PATP) Molecules

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**Figure S1.** (a) EDS spectrum of the Pt-based hollow nanostructures with atomic ratio of Co to Pt of 30:70. (b) Powder XRD patterns with various compositions. The XRD data illustrate the sample is face-centered cubic (fcc) structure of CoPt and provide evidence of the alloy structure with peak position shift.



Figure S2. EELs images of the Pt-based bimetallic hollow nanostructure.



**Figure S3.** HR-TEM images from the area of the Pt-based hollow nanostructure shown in Figure 2a. (a) and (b) show that the lattice inter-spacing of this Pt-based hollow nanostructure is 0.19 nm, corresponding to the Co metal. (c) and (d) show that the lattice inter-spacing of this Pt-based hollow nanostructure is 0.22 nm, corresponding to the Pt metal.



**Figure S4.** The emission spectrum of the Pt-based bimetallic hollow nanostructure with the laser wavelength 504 nm.



**Figure S5.** The SERS spectrum of PATP on the Pt-based hollow NC and Raman spectrum of solid PATP with the peak at ~1580 cm<sup>-1</sup>.

$$G = (I_{SERs}/N_{SERs})/(I_{bulk}/N_{bulk})$$
(1)

The number of tip molecules is represented as:

$$N_{SERs} = A \times N_{sub} \times A_{sub} / \sigma \tag{2}$$

where A is the laser irradiation area (~1.5  $\mu$ m),  $N_{sub}$  is the Pt-based hollow NC number density (~1.3×10<sup>-7</sup> nm<sup>-2</sup>) and the diameter size (~300 nm) of NC is calculated from Figure 1,  $A_{sub}$  is the area of single Pt-based hollow NC (~7.07×10<sup>4</sup> nm<sup>2</sup>),  $\sigma$  is adsorbed area of the single 4-MBA molecule (~0.2 nm<sup>2</sup>). It is assumed here that the substrate material surface is fully adsorbed with 4-MBA molecules and one layer of Pt-based hollow NC was deposited on the substrate homogeneously.

The number of tip molecules contributing Raman signal in the experiment is:

$$N_{\text{bulk}} = A \times h \times n_{\text{bulk}} = A \times h \times N_{\text{A}} \times \rho_{\text{bulk}} / M_{\text{bulk}}$$
(3)

where A is the laser irradiation area (~1.5  $\mu$ m), h is the laser focusing depth (~15  $\mu$ m), n<sub>bulk</sub> is the number of tip molecules per unit volume,  $\rho_{bulk}$  is the density of bulk (~21.45 g/cm<sup>3</sup>), M<sub>bulk</sub> is the molecular weight of PATP molecule (~125.19 g/mol), N<sub>A</sub> is the Avogadro constant (~6.02×10<sup>23</sup>).

It can be calculated as:

$$G = (I_{SERs}/N_{SERs})/(I_{bulk}/N_{bulk}) = (I_{SERs}/I_{bulk}) \times (\sigma \times h \times \rho_{bulk} \times N_A)/(N_{sub} \times A_{sub} \times M_{bulk})$$

$$(4)$$

So the EF factor G is approximately calculated as 1×10<sup>6</sup>.