

Supplementary Information for

Graphene-interlayered magnetic composite as a multifunctional SERS substrate

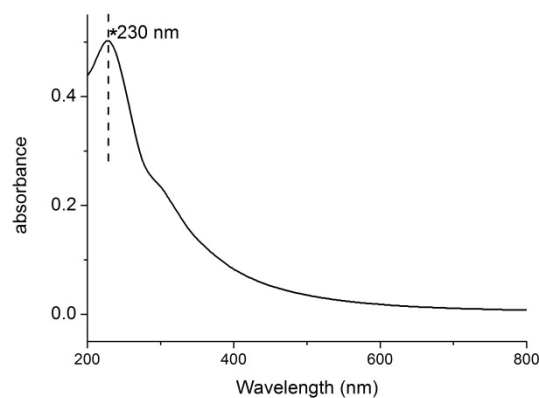


Figure. S1 UV-vis of GO nanosheets solution

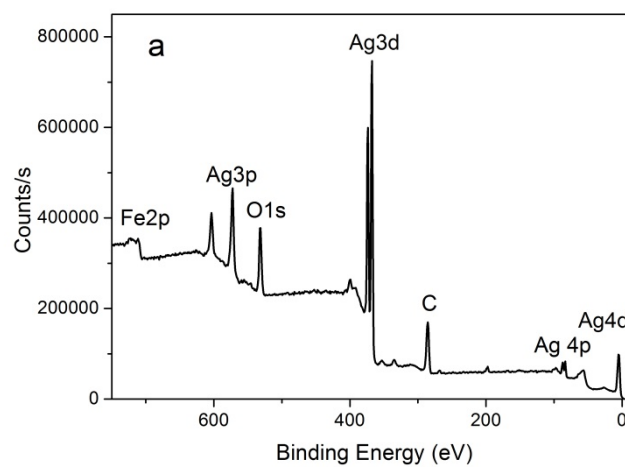


Figure. S2 (a). XPS survey spectrum of $\text{Fe}_3\text{O}_4@\text{Ag-GO}$

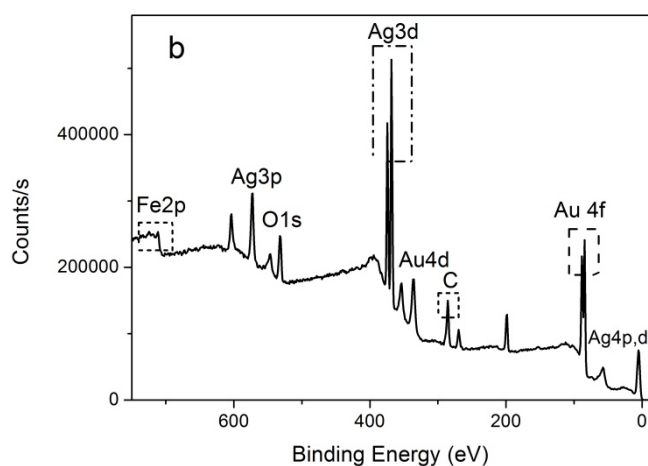


Figure. S2 (b). XPS survey spectrum of $\text{Fe}_3\text{O}_4@\text{Ag-rGO-Au}$. Elements labeled with dash rectangle were analysed in detail in the body.

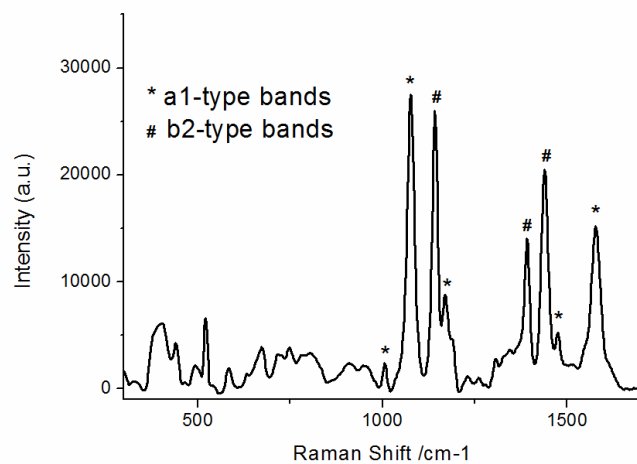


Figure. S3 SERS spectrum of PATP at a wavelength of 782 nm

Normal Raman	SERS Signal	assignments
	In 785 nm	
391	--	$\delta\text{CS},(a_1)$
463	--	$\gamma\text{CCC},7a(a_1)$
634	--	$\gamma\text{CCC},12(a_1)$
1004	1005	$\delta\text{CC} + \gamma\text{CCC},18a(a_1)$
1076	1076	$\nu\text{CS},7a(a_1)$
1140	1142	$\delta\text{CH},9b(b_2)$
1172	1172	$\delta\text{CH},9a(a_1)$
	1392	$\nu\text{CS}_+ \delta\text{CH},14b(b_2)$
	1439	$\nu\text{CS}_+ \delta\text{CH},(3b_2)$
1482	1475	$\nu\text{CS}_+ \delta\text{CH},19a(a_1)$

Table S1. Raman peaks of PATP and according assignments**EF calculation:**

The SERS enhancement factors (EF) for PATP can be calculated according to the equation $EF = (I_{SERS}/I_{bulk})(N_{bulk}/N_{SERS})$, where I_{bulk} is the intensity of Raman signal as calculated from the target peak area and N_{bulk} is the number of molecules in the focal volume, whereas I_{SERS} and N_{SERS} are parameters when $Fe_3O_4@Ag-rGO-Au$ microspheres are used as active substrates. For valuable determination of N_{bulk} and N_{SERS} , 20 ul melted PATP sample and the PATP-absorbed $Fe_3O_4@Ag-rGO-Au$ microspheres (separated from PATP solution 10^{-7} M, 2 ml) were uniformly spread on 300 nm SiO_2/Si substrates.

In detail, The solid sample of PATP was slowly heated to 42 °C and then the melted PATP (20 ul) was dropped on 0.4 cm×0.4cm SiO_2/Si substrate, formed a uniform film (~800 um). After totally cooled down, the SERS detection was conducted and the peak intensity of acquired Raman signal was I_{bulk} . The penetration

depth of laser (h) is calculated by equation: $h = \int_{-\infty}^{\infty} I(z)dz/I_{max}$, $I(z)$ is the intensity of the Raman peak of Si (520 cm^{-1}), which is measured as a reference to calculate h. As displayed in Fig. S4c, it is a function of the distance (z) deviated from the focused center. According to the equation, the penetration depth of laser (h) is measured as 460 μm. Therefore, $N_{bulk} = \pi(d/2)^2h\rho_0N_A/M_0 = 5.12 \times 10^{14}$. ρ_0 (1.18 g/mL) and M_0 (125.19 g/mol) is the density and molar mass of melted PATP, respectively.

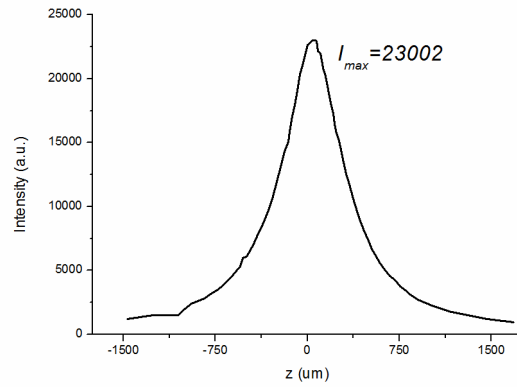


Figure. S4 Intensity of the Raman peak of Si (520 cm^{-1}) $I(z)$, a function of the distance (z) deviated from the focused center.

The number of PATP molecules contribute to the SERS signals within the laser spot area is $N_{\text{SERS}} = RA/\sigma$, under the assumption that molecules were absorbed on the microspheres in single layer uniformly. R is the roughness factor of the nanostructure, which approximate to be 1. σ is the occupied surface area of a single molecule, which is equal to 0.22 nm^2 for PATP. A is the area of laser spot, $A = \pi(d/2)^2$. Therefore, $N_{\text{SERS}} = \pi(d/2)^2/\sigma$. The diameter of spot size d is $105\mu\text{m}$. Finally, the $N_{\text{bulk}}/N_{\text{SERS}} = h\rho_0\sigma N_A/M_0$, and is calculated equal to 5.61×10^5 .

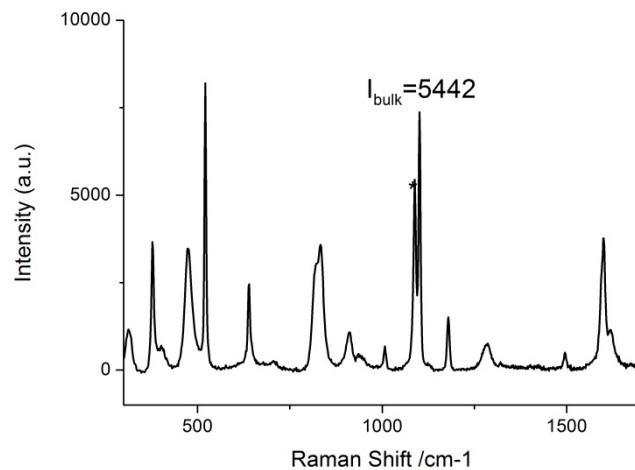


Figure. S5 (a) Raman spectrum of pure PATP

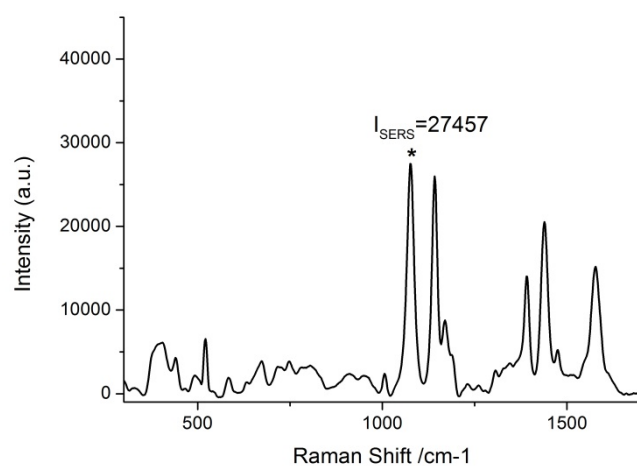


Figure. 5 (b) SERS spectrum of PATP with a concentration of 10^{-7}M

The $I_{\text{SERS}}/I_{\text{bulk}}$ was $27457/5442=5.04$ for peaks at 1077 cm^{-1} , reading from the spectra presented in Fig. S5 a and b. Accordingly, the EF value of peaks at 1077 cm^{-1} is calculated to be about 2.83×10^6 .