# Revealing the role of interfacial properties on catalytic behaviors by in situ surface-enhanced Raman spectroscopy

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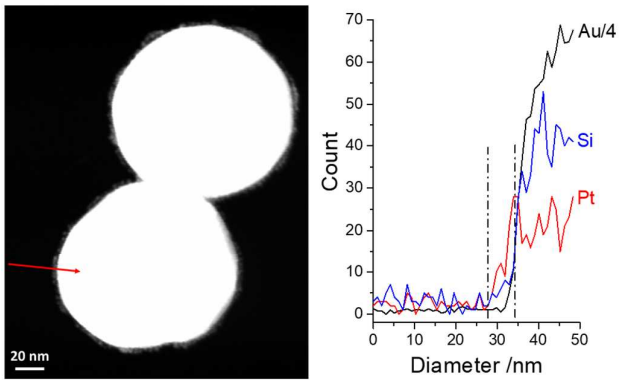


**Figure S1.** elemental line scan analysis of Pt-on-SHINs.

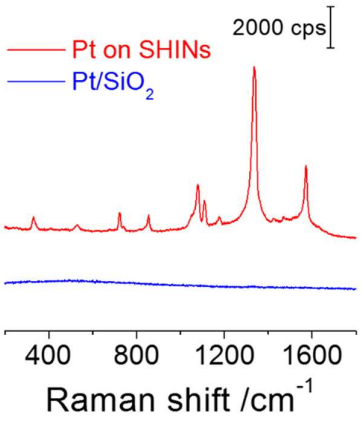


**Figure S2.** Raman spectra of pNTP adsorption on Pt-on-SHINs and $Pt/SiO_2$

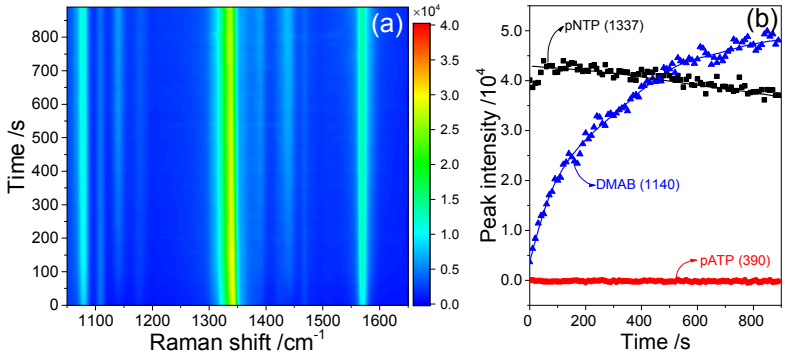


**Figure S3.** *In-situ* SERS spectra of the hydrogenation of pNTP on SHINs (a) and the corresponding intensity of pNTP, DMAB, and pATP as a function of reaction time (b).

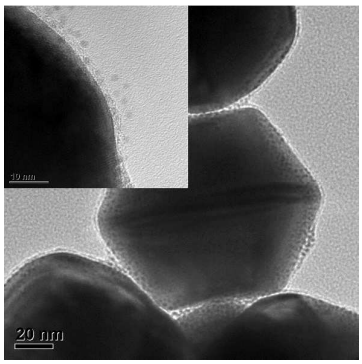


**Figure S4.** HR-TEM images of Pt-on-SHINs after reaction.

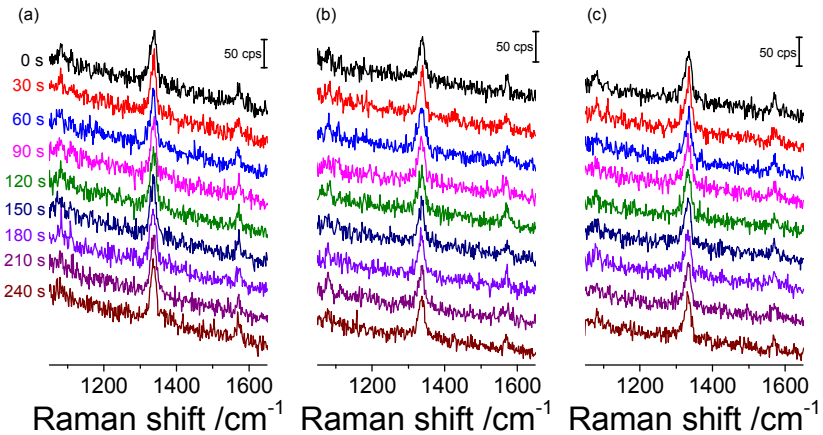


**Figure S5.** In-situ SERS spectra of the conversion of pNTP on pinhole-free SHINs under different conditions. (a) 25 $^{o}$C, Ar. (b) 60 $^{o}$C, Ar. (c) 60 $^{o}$C, $H_2$. The intensities of the Raman signals for pNTP on the pinhole-free SHINs are much lower than those on Pt-on-SHINs (pinhole-free) due to the weak adsorption of pNTP on silica.

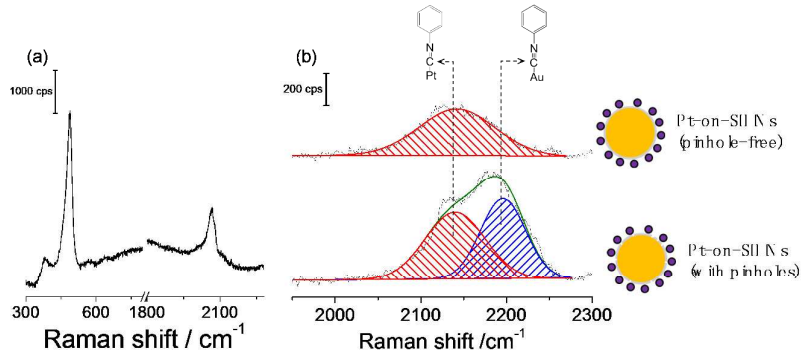


**Figure S6.** In situ SHINERS spectra of CO (a) and phenyl isocyanide (b) adsorbed on Pt-on-SHINs (pinhole-free)

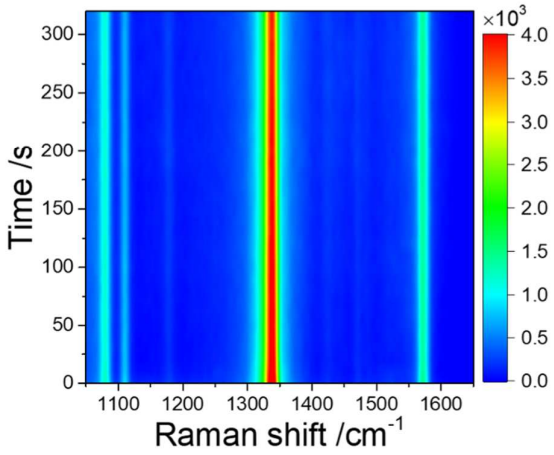


**Figure S7.** In-situ SERS spectra of the conversion of pNTP on Pt-on-SHINs (pinhole-free) under Ar at 60 °C.

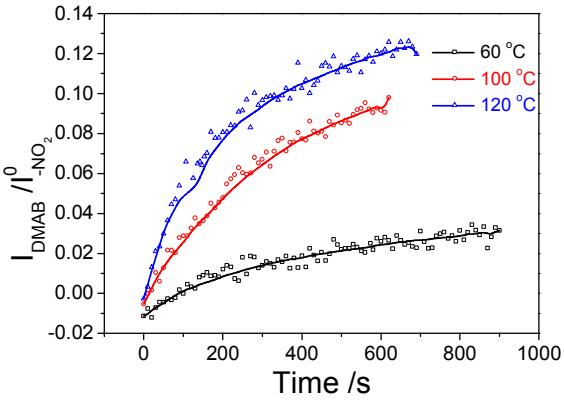


**Figure S8.** Effect of temperature on the yield of DMAB on Pt-on-Au via the photo-induced coupling reaction under Ar.

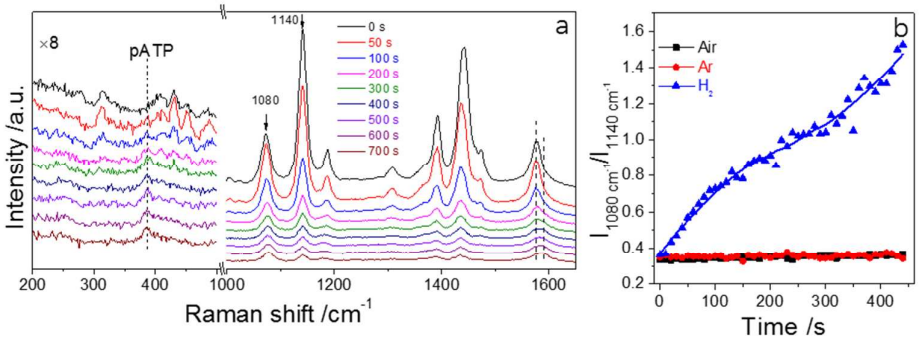


**Figure S9.** (a) In situ SERS spectra of the hydrogenation of DMAB on Pt-on-Au. It can be clearly seen that DMAB can be efficiently converted to pATP. (b) The ratio of the Raman band at 1080 $cm^{-1}$ to that at 1140 $cm^{-1}$ under different gaseous atmosphere as a function of time. Both DMAB and pATP show a strong Raman band at 1080 $cm^{-1}$, but only DMAB show a Raman band at 1140 $cm^{-1}$. Thus, the formation of pATP can also be speculated based on the ratio of the Raman band at 1080 $cm^{-1}$ to that at 1140 $cm^{-1}$. This ratio only increases under $H_2$, and maintains unchanged under Ar and air. It indicates the conversion of DMAB to pATP is triggered by $H_2$ rather than the laser.

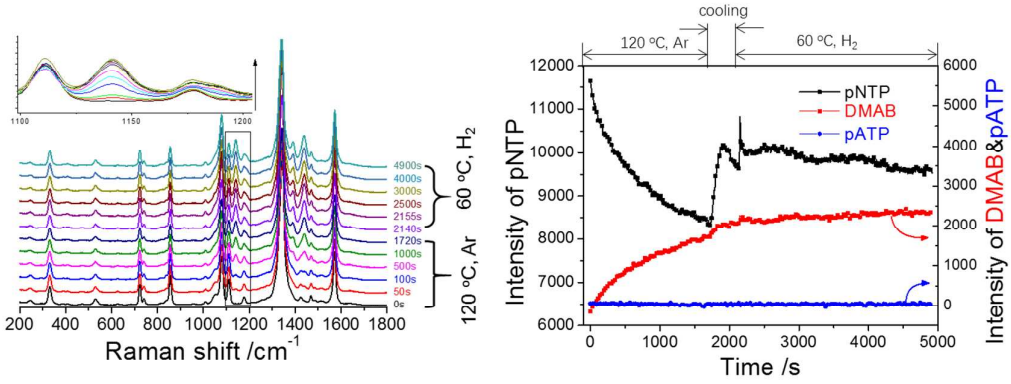


**Figure S10.** In situ SERS spectra of pNTP adsorbed on bare Au nanoparticles under Ar and $H_2$.

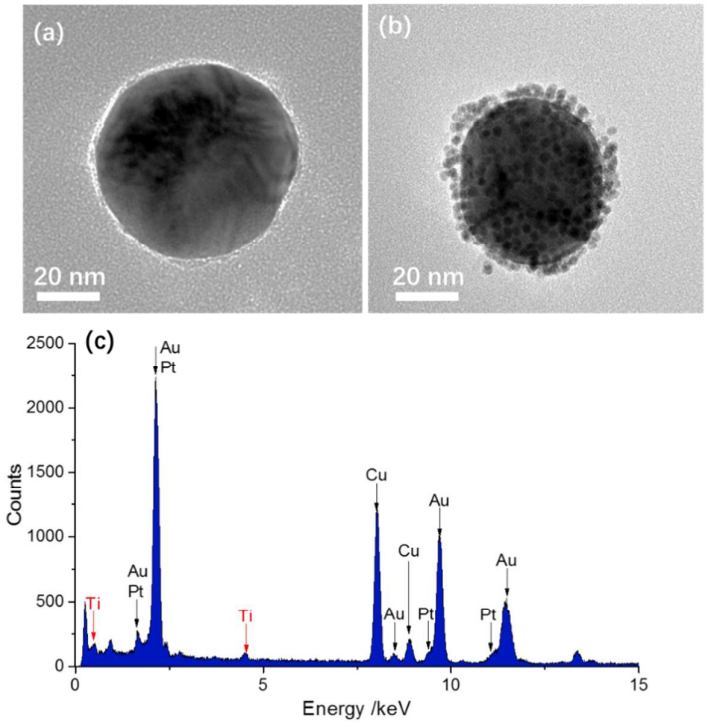


**Figure S11.** TEM image of SHINs with a $TiO_2$ shell (a) and the nanocomposite of Pt nanocatalysts on this kind of SHINs (Pt-on-SHINs ($TiO_2$ shell)) (b). (c) Energy dispersive spectroscopy (EDS) of the nanocomposites. Pt nanocatalysts are highly dispersed on the surface of the SHINs. The EDS analysis demonstrates that the shells are composed of $TiO_2$.