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1 The Charge Transfer Effect on SERS in a Gold-decorated Surface Defect

2 Anatase Nanosheet/ Methylene Blue (MB) system

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15 **S1** The morphology of anatase nanosheets on silicon wafers



500 nm

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 $\label{eq:supplementary Figure 1} SEM \ images \ of \ TiO_2, \ TiO_2(D) \ and \ TiO_2(D)-Au \ on \ silicon \ wafers.$

19 The EF was calculated following the formula^[s1]:</sup>

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$$EF = (I_{SERS}/N_{SERS})/(I_{bulk}/N_{bulk})$$
(1)

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$$N_{SERS} = CVN_A A_{Raman} / A_{sub}$$
(2)

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$$N_{bulk} = \rho h N_A A_{Raman} / M \tag{3}$$

Here, I_{SERS} and I_{bulk} were the intensities of the selected Raman Peak in SERS (Supplementary Figure 2) and non-SERS spectra (Supplementary Figure 3), and N_{SERS} and N_{bulk} were the average number of MB molecules in the scattering area for SERS and non-SERS measurement. The intensities of Raman were obtained by taking average of 10 spots on one sample. The I_{bulk} as reference was got using MB (0.05 M) dispersed on Si wafer at the same condition following SERS sample preparation. The N_{SERS} can be estimated using equation (2) with the molar concentration of the analyte solution (*C*), volume of the droplet (*V*), Avogadro constant (N_A) and the laser spot area (A_{Raman} , 1 µm in diameter). The N_{bulk} can be calculated using equation (3). The confocal depth (*h*) of the laser beam is 21 μ m and M is the molecular weight, ρ is the density of bulk MB.



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37 Reference

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- S1. Cong, S.;Yuan, Y.;Chen, Z.;Hou, J.;Yang, M.;Su, Y.;Zhang, Y.;Li, L.;Li, Q.; Geng, F. Noble metal-comparable
 SERS enhancement from semiconducting metal oxides by making oxygen vacancies. *Nature communications* 2015, 6, 1-7.

each sample.

42