

## Electronic Supporting Information

### Plasmon Enhanced Strong Visible Light Photocatalysis by Defect Engineered CVD Graphene and Graphene Oxide Physically Functionalized with Au Nanoparticles

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#### ***SI1: Preparation of Graphene and Graphene Oxide***

First, a Cu foil (purity: 99.8 %; Alfa–Aesar) was inserted into a quartz chamber and purged with argon gas for 5 min. Then, the temperature was increased to 1000°C (growth temperature of graphene) at the rate of 16°C min<sup>-1</sup> using a horizontal muffle furnace (Indfur, India). The chamber pressure was maintained at high vacuum (1x10<sup>-4</sup> mbar) during the heating. The Cu foil was pre-annealed at the same temperature in H<sub>2</sub> gas environment (200 sccm (standard cubic centimetres per minute) flow rate) for 30 min in order to increase the Cu grain size as well as to avoid the oxidation of the Cu surface. The graphene growth was carried out by controlled flow of CH<sub>4</sub> ~18 sccm and H<sub>2</sub> ~ 200 sccm for 30 min at a temperature of 1000°C and a pressure of 4.0 mbar. Since graphene was deposited on both sides of the Cu foil, the bottom graphene was removed by diamond polishing. Graphene layer on top of the Cu foil was transferred on to Si/SiO<sub>2</sub>, quartz and TEM Cu grid by a conventional wet transfer technique.[45] In this process, Poly (methyl methacrylate) (PMMA)/Toluene was spin coated on the graphene/Cu and the underlying Cu was etched in an aqueous Fe (NO<sub>3</sub>)<sub>3</sub> etchant solution. PMMA/graphene, floating on the etchant solution was rinsed in deionised water for a few times, until the PMMA and

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metal impurities were removed. Then it was scooped and transferred on to the required substrates.

## **SI2. H<sub>2</sub> Annealing of Graphene**

Clean transfer of graphene from Cu foil on to the required substrates is one of the challenging tasks. Though wet transfer is a conventional protocol, there are still leftover impurities (PMMA and catalyst NPs along with the oxide traces) present on the graphene substrate. In order to remove these impurities from the graphene on Si/SiO<sub>2</sub> and TEM grid, these samples were annealed in H<sub>2</sub> atmosphere. Initially, the samples were taken in a quartz boat and kept in the tubular chamber. The chamber was pumped up to a base vacuum of 10<sup>-5</sup> mbar followed by ramping of the temperature up to 400°C. The graphene annealing was performed under H<sub>2</sub> gas environment for 2 hours. The annealing parameters were chosen optimum to achieve a clean graphene without the residual impurities on the graphene surface.[46] The degree of improvement in the crystallinity and spatial homogeneity of graphene layers was confirmed by Raman spectra and Raman mapping, respectively, in each case before and after annealing.

## **SI3: Preparation of Graphene Oxide**

Graphene oxide (GO) was prepared by the modified Hummers method. Initially, 10 g of high purity (99.98%) bulk graphite (Average flake area ~ 500 μm<sup>2</sup>, ASBURY CARBON, USA, carbon content > 98 wt%) and 5 g of NaNO<sub>3</sub> were mixed with sulphuric acid (230 ml, 98 wt%) in an ice bath, and 30 g of KMnO<sub>4</sub> was slowly added to prevent the temperature from exceeding 293 K. The reaction was conducted at 308 K for 30 min with gas release, and then de-ionized water (460 ml) was gradually added, bringing about violent effervescence. The temperature of the water bath was increased to 371 K and the reaction was maintained for 40 min in order to increase the oxidation degree of the GO product. The resultant bright–yellow suspension was

diluted and further treated with a  $\text{H}_2\text{O}_2$  solution (30 ml, 30%), followed by centrifugation and careful washing to clean out the residual salt. The wet GO was dewatered by vacuum drying at 323 K.

A GO suspension in water (2mg /ml, 200 ml) was treated in an ultrasonic cleaner (KQ-100, 40 kHz, 100 W) for 30 min, followed by high-speed centrifugation (10000 rpm, 20 min) to remove impurities, which only resulted in a slight precipitation. The stable GO hydrosol was heated at 353 K for a short period (around 20 min for a 5 mm thick membrane and 40 min for a 10mm thick membrane) in a thermostatic water bath; during this time a smooth and condensed thin film was formed very rapidly at the liquid/air interface. The under surface suspension was then transferred into another beaker, and the membrane left at the bottom was dried at 353 K for 8 h and torn off.

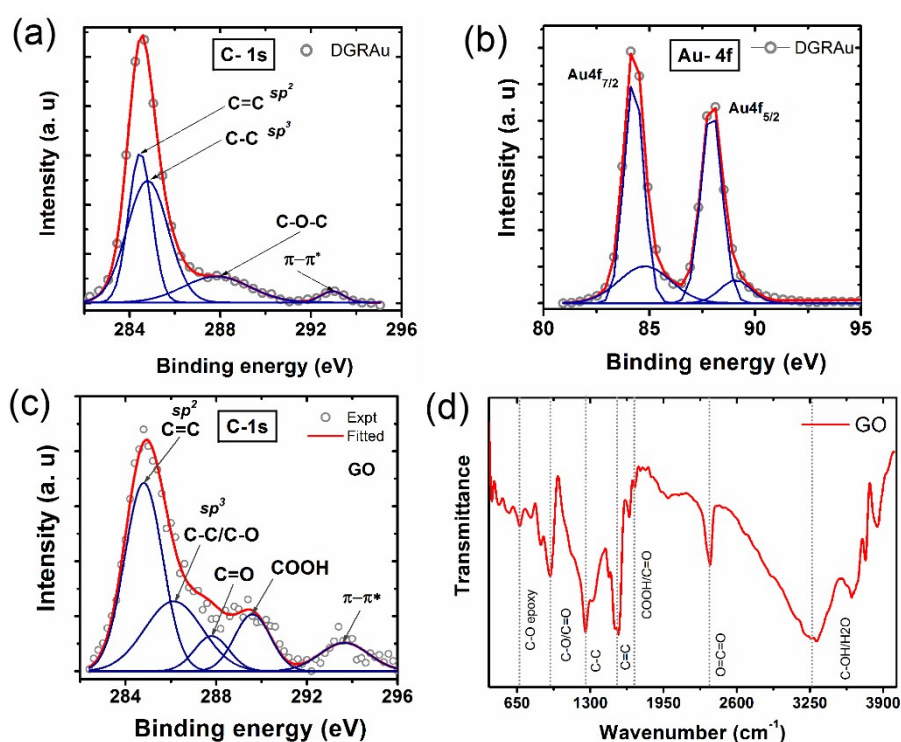


Figure S1: XPS spectra for the core level peaks of (a) C-1s, (b) Au-4f in DGRAu sample. (c) XPS spectra for the core level peak of C-1s in GO. Chemical bonding of carbon and oxygen functional groups are identified from the Gaussian line shape. (d) FTIR spectra of GO. The vertical lines of each band indicate the chemical functional group in GO.

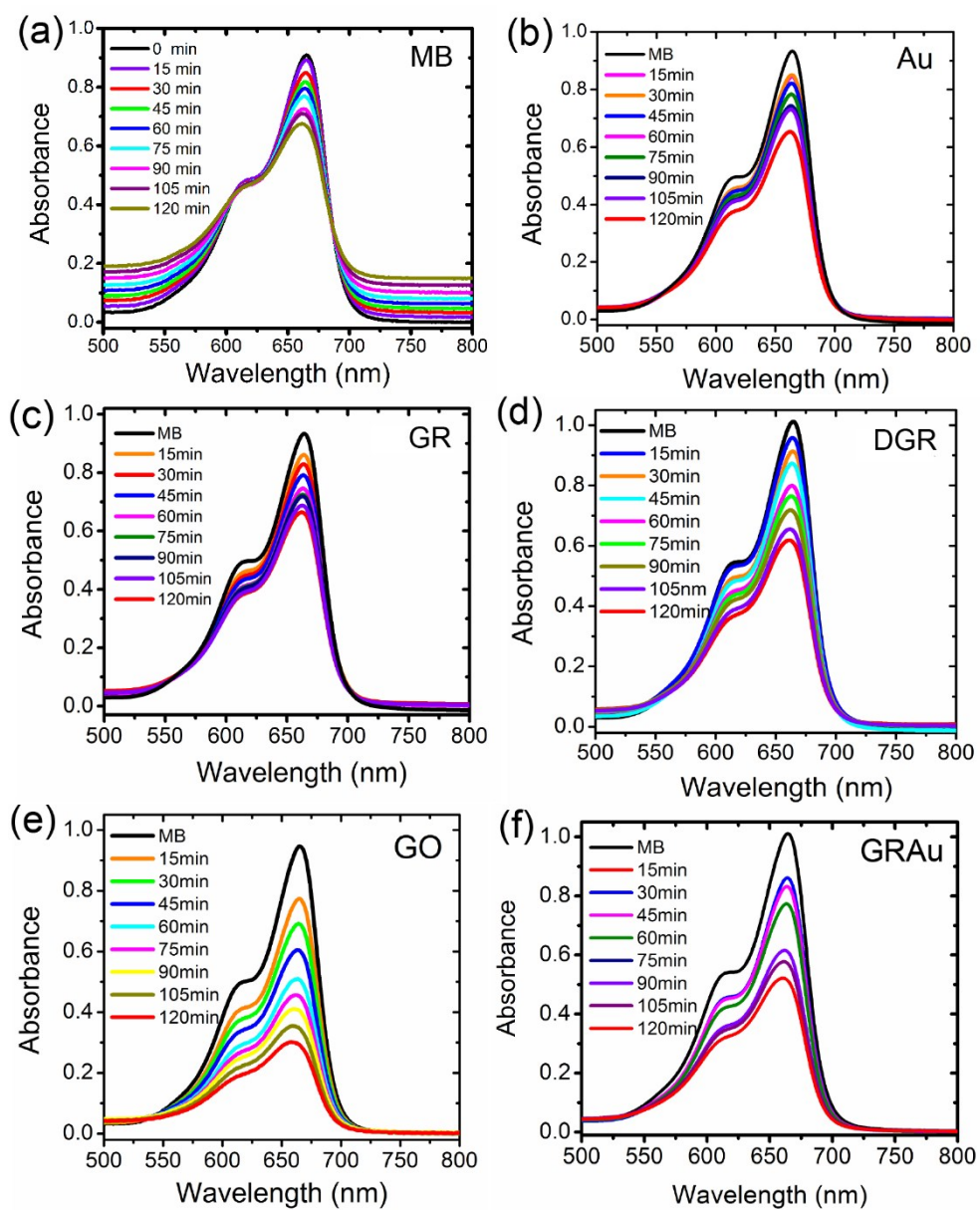


Figure S2: Evolution of the absorption spectra showing visible light photocatalytic degradation performance of various graphene-plasmonic catalyst substrates in a standard MB aqueous dye solution: (a) MB, (b) Au, (c) GR, (d) DGR, (e) GO, (f) GRAu.