Supplementary information

Facile synthetic route to exfoliate high quality and super-large lateral size of graphene-based sheets and their applications in SERS and CO₂ gas sensing

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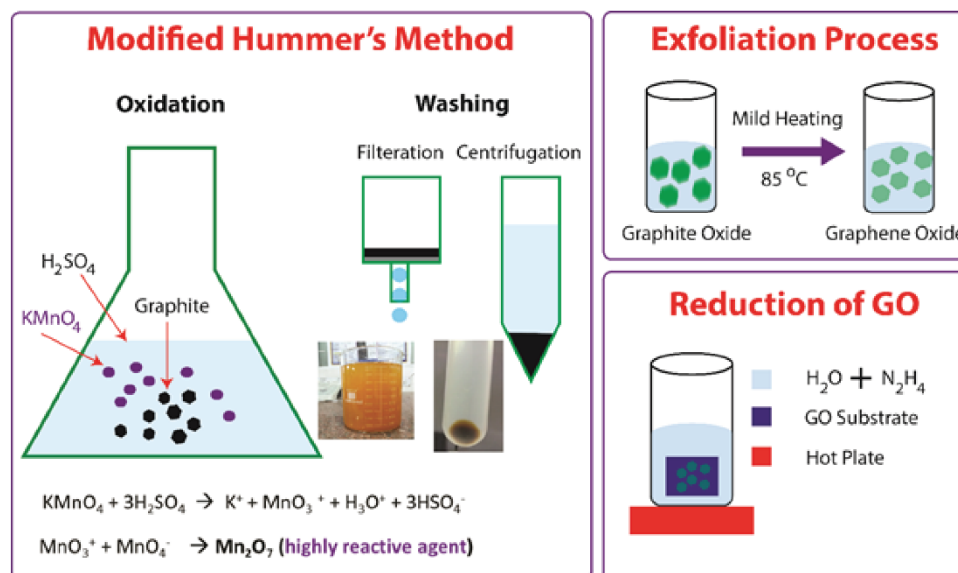Email address: giri@iitg.ac.in (P. K. Giri)

Fig. S1. Schematic illustration of the synthesis of super large area GO, exfoliation using mild heating, and chemical reduction to develop super large area rGO.
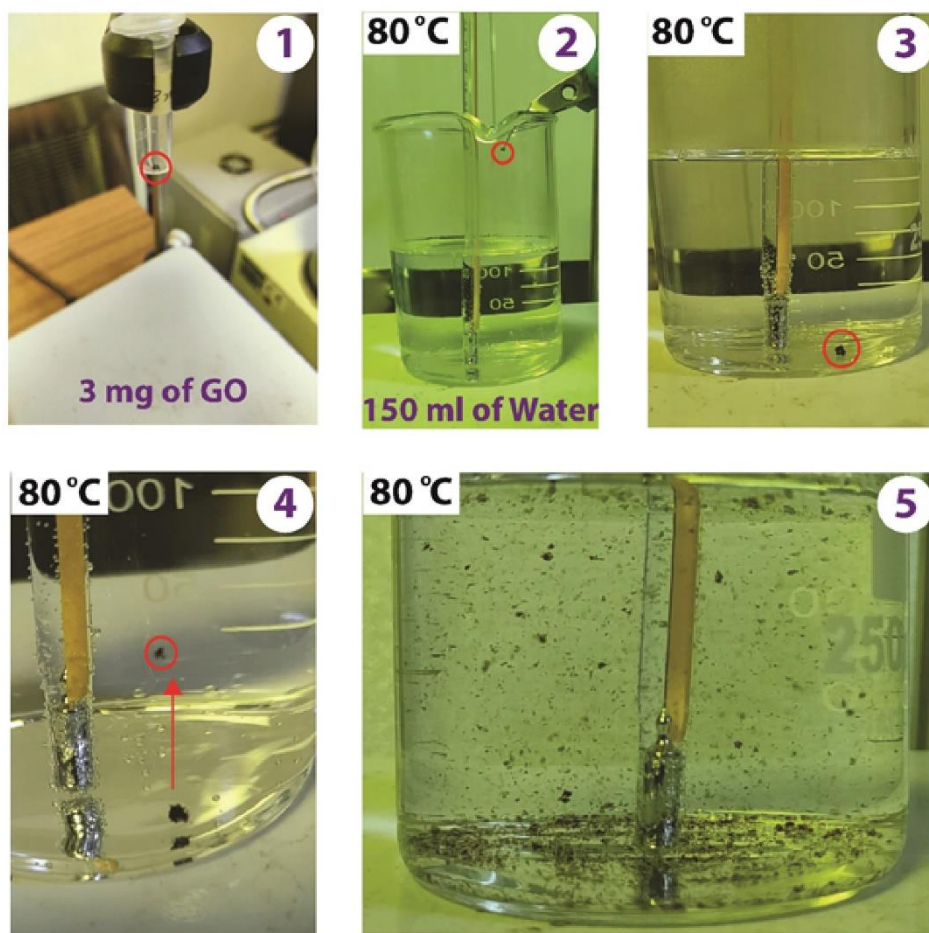
Fig. S2. Snapshots showing stages of exfoliation during mild heating condition. The temperature is carefully adjusted to maintain a constant value. The rise of initial exfoliated flake is observed at stage 4 driven by mild temperature. At stage 5, numerous fragmented flakes assist in mechanical exfoliation within the flakes owing to the gained kinetic energy.
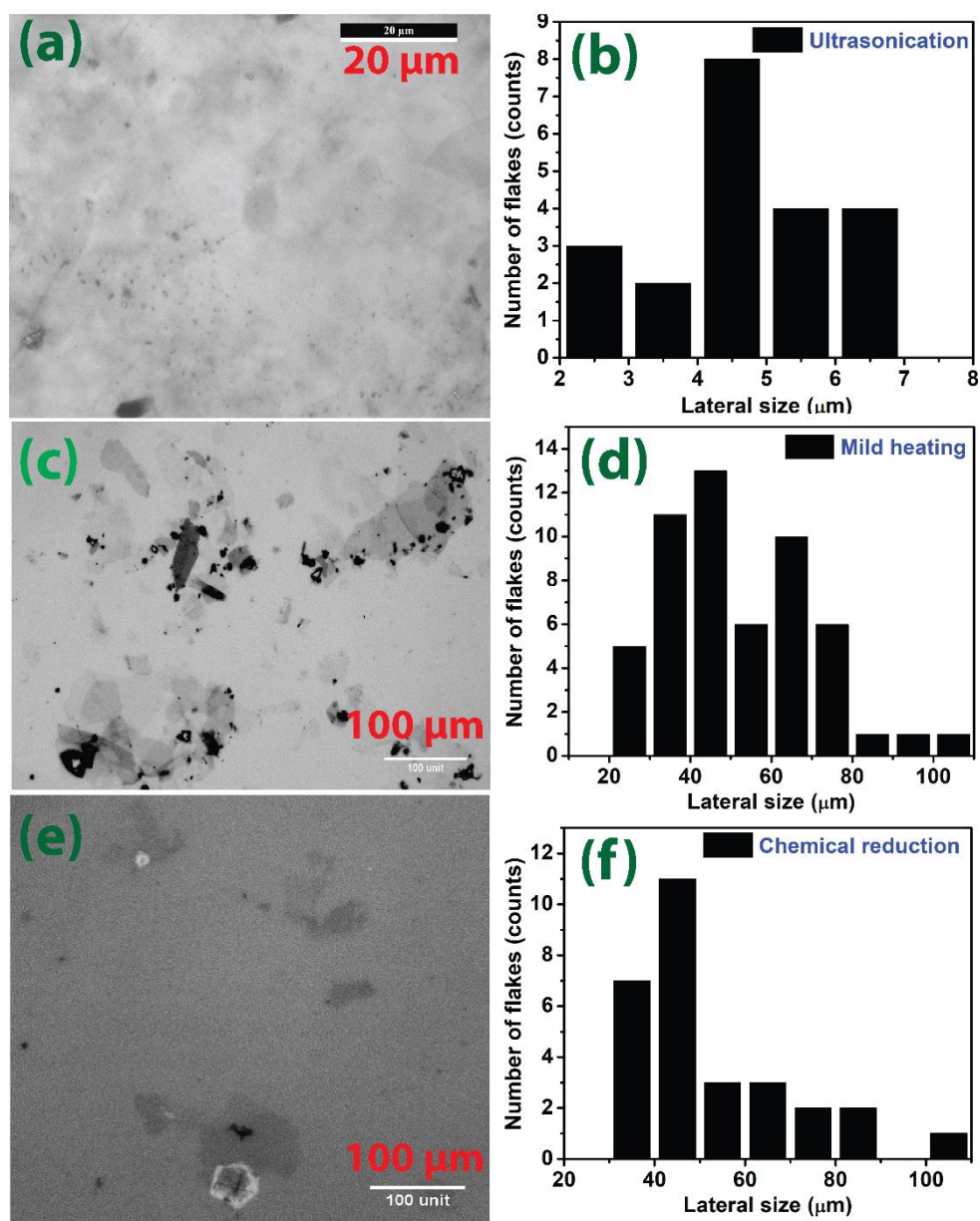
Fig. S3. Optical images showing various lateral sizes for (a) GO from ultrasonic exfoliation (UGO), (c) GO from mild heating exfoliation (GO0, and (e) rGO from chemical reduction. Histogram depicting statistical distribution of various lateral sizes for (b) GO exfoliation from ultrasonication, (d) GO exfoliation from mild heating, and (e) rGO from chemical reduction.
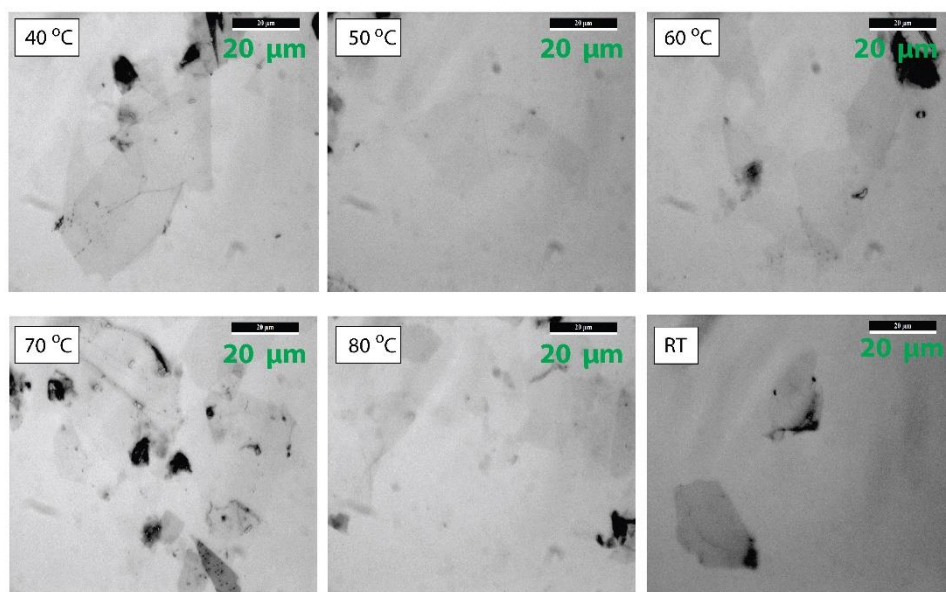
Fig. S4. Optical images of exfoliated GO obtained from mild heating at different temperatures (RT stands for room temperature, 23 °C). Darker patches correspond to the thicker sheets, which are not efficiently exfoliated.
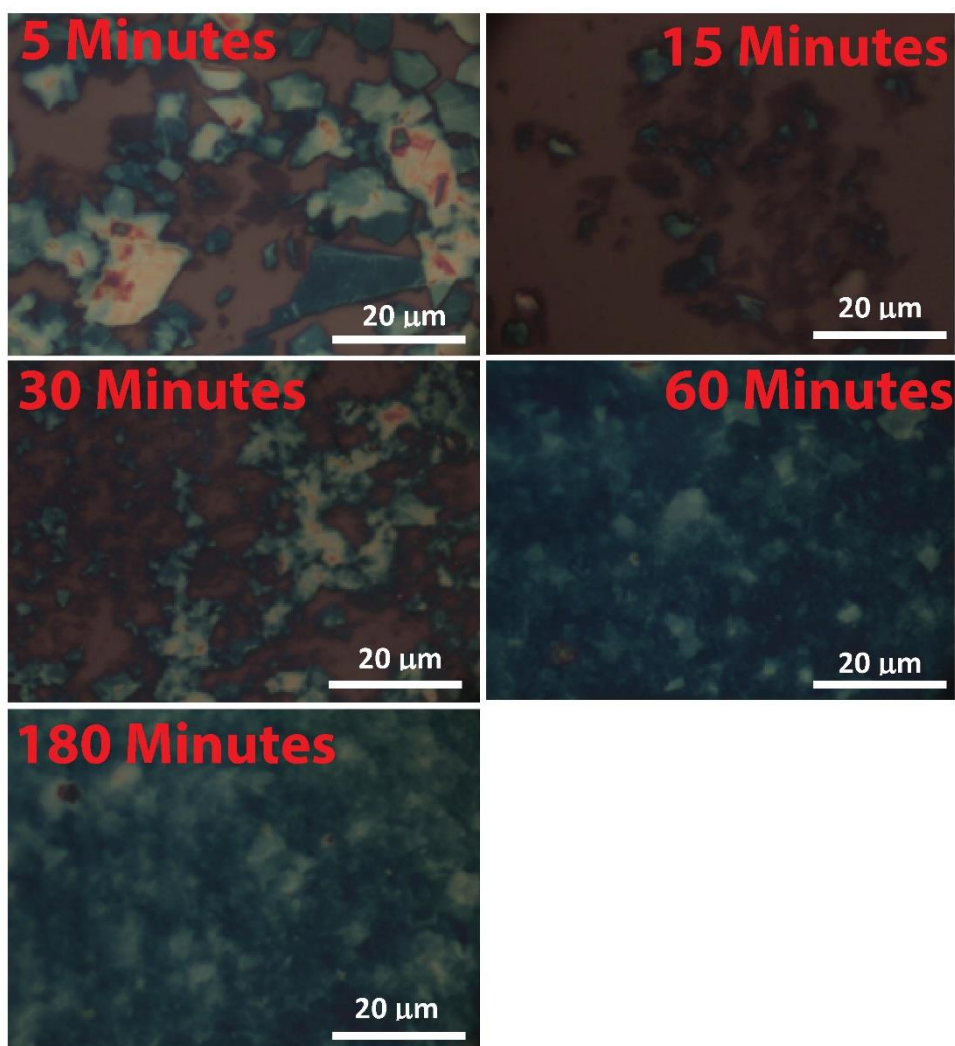
Fig. S5. Optical images of GO flakes obtained from exfoliation using ultrasonication at different time intervals at 50 W. Flakes fragmentation and layer exfoliation occur simultaneously wherein fragmentation predominates over exfoliation as indicated by the presence of several smaller lateral size flakes.
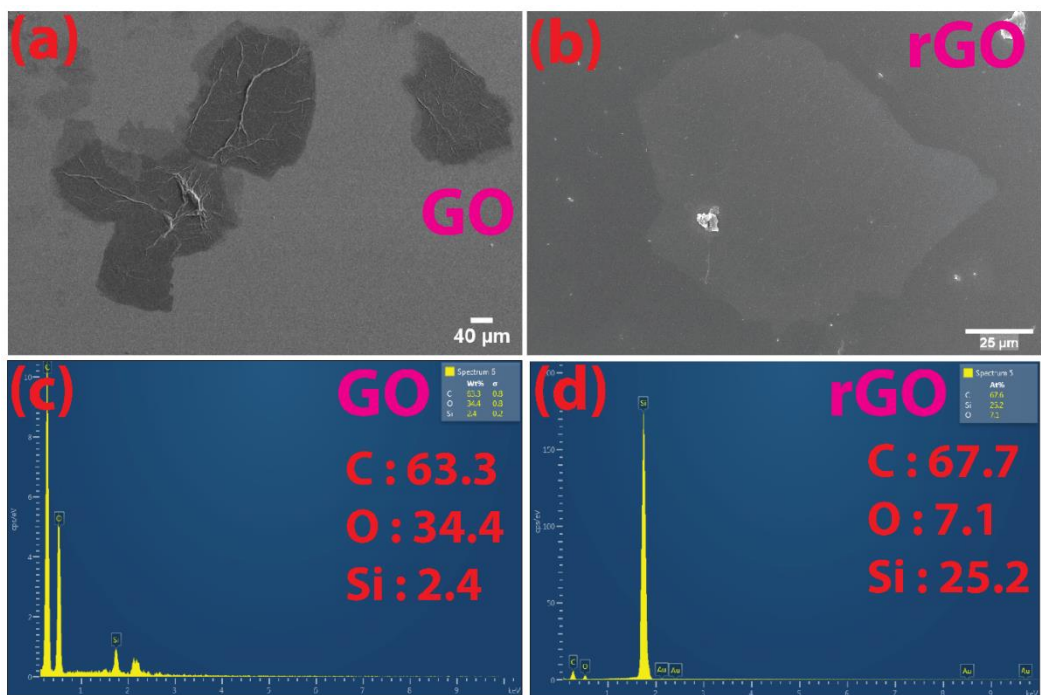
Fig. S6. (a) FESEM image of (a)the GO, (b) the rGO sheets; EDX elemental analysis of (c) the GO, and (d) rGO sheets.
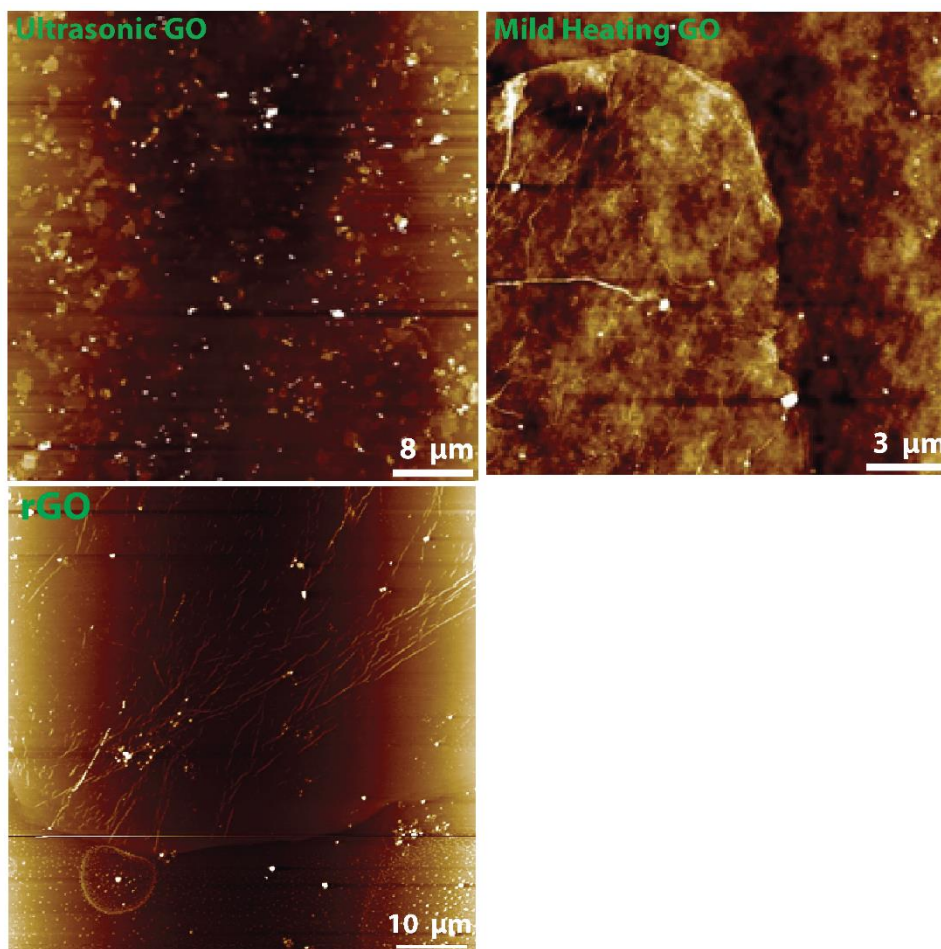
Fig. S7. Lower magnification AFM micrographs showing exfoliated flakes obtained using (a) ultrasonication for GO, (b) mild heating for GO and (c) chemically reduced rGO.
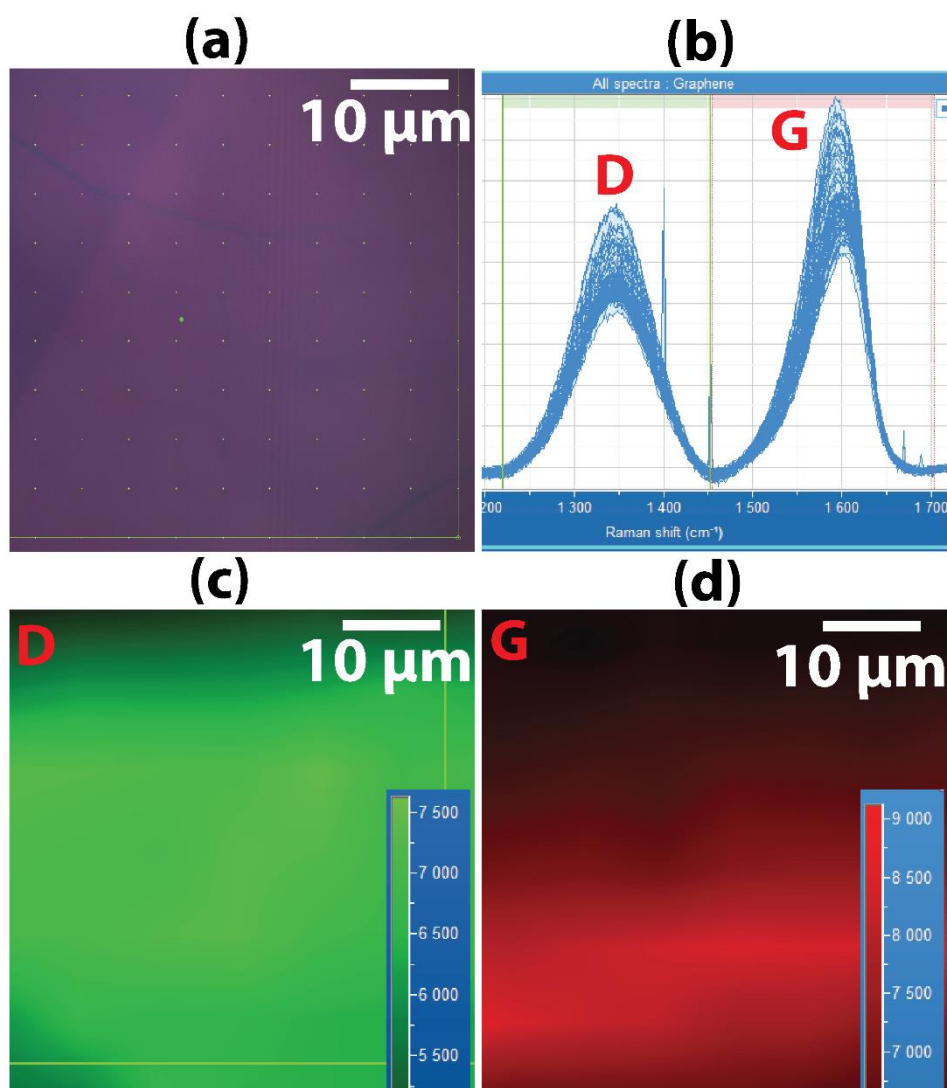
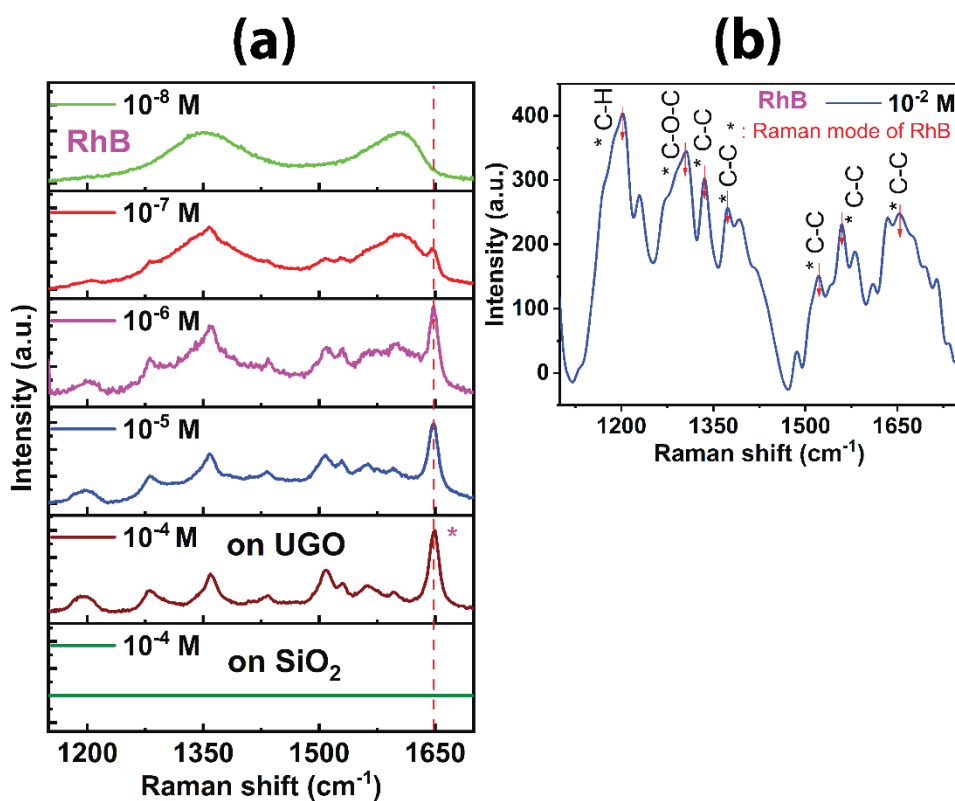**Fig. S8**. Raman mapping spectrum on the 50 x 50 µm size rGO sheet.

Fig. S9. (a) Raman spectra of different concentrations of RhB on UGO; (b)Raman spectrum of 10^{-2} M concentration of RhB on SiO$_2$/Si substrate.

Table S1. Raman peak positions for RhB molecule[1]

Raman Peak Position (cm^{-1})	Bond details
1647	C-C stretching
1595	C-C stretching
1560	C-C stretching
1508	C-C stretching
1358	C-C stretching
1281	C-O-C stretching
1199	C-H Bending

During SERS technique, if different substrate samples are used with different Raman characterization parameters such as laser power and accumulation time then the formula for the enhancement factor is below,

$$EF = \frac{I_{SERS}}{I_R} \times \frac{C_R}{C_{SERS}} \times \frac{P_R}{P_{SERS}} \times \frac{T_R}{T_{SERS}}$$

Whereas, P_{SERS} is the laser excitation power in SERS measurement on GO/rGO substrate, T_{SERS} is the accumulation time of the SERS measurement on GO/rGO substrate, P_{R} is the laser excitation power of the Raman signal on base SiO_2 substrate, and T_{R} is the accumulation time of the Raman signal on base SiO_2 substrate.

If the volume of the sample solution is used differently in Raman characterization measurement, then the EF formula[2] is given below,

$$EF = \frac{I_{SERS}}{I_R} \times \frac{C_R}{C_{SERS}} \times \frac{P_R}{P_{SERS}} \times \frac{T_R}{T_{SERS}} \times \frac{N_R}{N_{SERS}}$$

Whereas, N_{SERS} is the volume of the concentration solution used for SERS signal on GO/rGO substrate, and N_{R} is the volume of the concentration solution used for the Raman signal on base SiO_2 substrate.

Table S2. Raman intensity and Enhancement Factors for various substrates at different Raman shifts

Peak position (cm^{-1})	Raman intensity for RhB on SiO_2 substrate (a.u.)	Raman intensity for RhB on rGO substrate (a.u.)	EF of rGO substrate	Raman intensity for RhB on GO substrate (a.u.)	EF of GO substrate
1647	247	7750	3137	23423	9482
1596	188	3261	1734	8571	4559
1505	150	4829	3219	12946	8630
1356	255	4806	1884	12158	4767
1286	344	3042	884	6642	1930
1196	403	1474	365	4277	790

Reference

1   S. Huh, J. Park, Y. S. Kim, K. S. Kim, B. H. Hong and J. M. Nam, *ACS Nano*, 2011, **5**, 9799–9806.

2   Y. Xu, M. P. Konrad, J. L. Trotter, C. P. McCoy and S. E. J. Bell, *Small*, , DOI:10.1002/smll.201602163.