Electronic Supplementary Material (ESI) for Nanoscale.

Excitation Wavelength Dependent Fluorescence of Graphene Oxide Controlled by Strain

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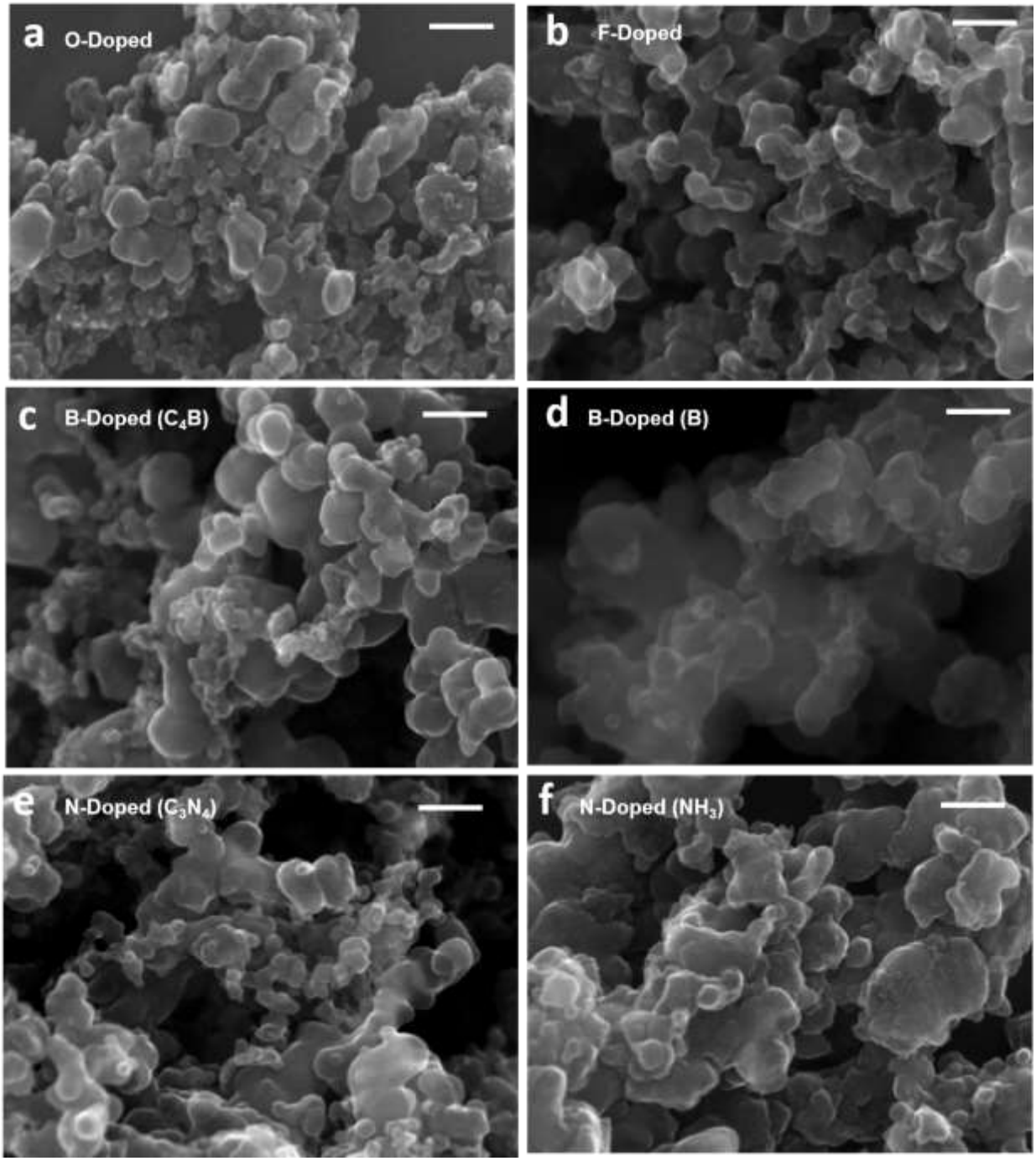

Figure S1. SEM Images of Doped GO. Scale bar is 100 nm for **a** O-doped GO, **b** F-doped GO, **c** C_4B B-doped GO, **d** pure B B-doped GO, **e** C_3N_4 N-doped GO, and **f** NH_3 N-doped GO.

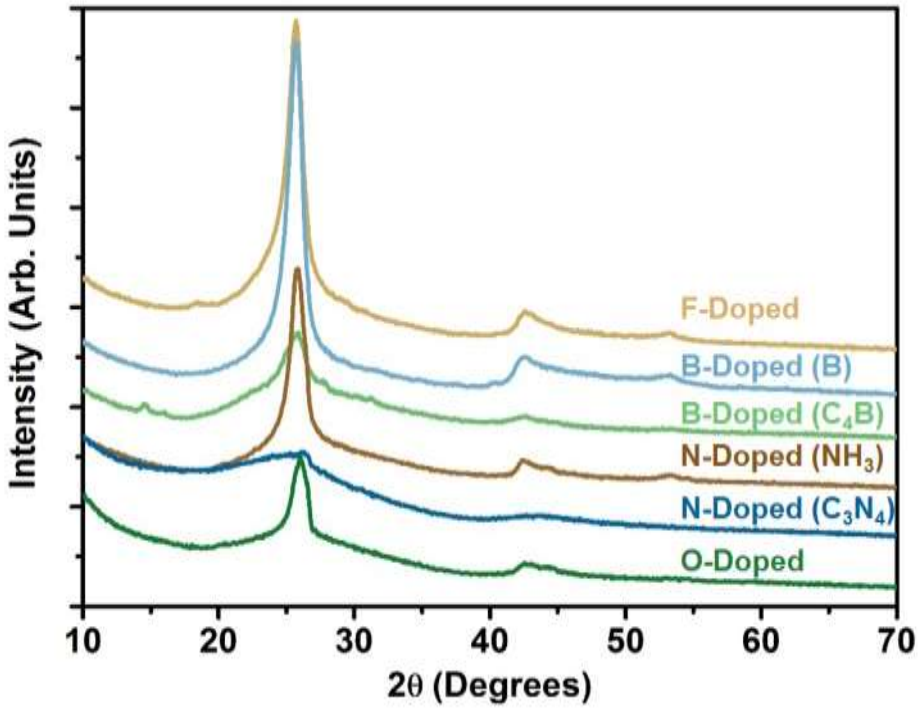

Figure S2. XRD patterns of doped graphene oxide samples. The peaks at $2\theta=26.6^{o}$, 42.5^{o}, 45.6^{o} and 54.8^{o} correspond to (002), (100), (101) and (004) phases of graphite, respectively.

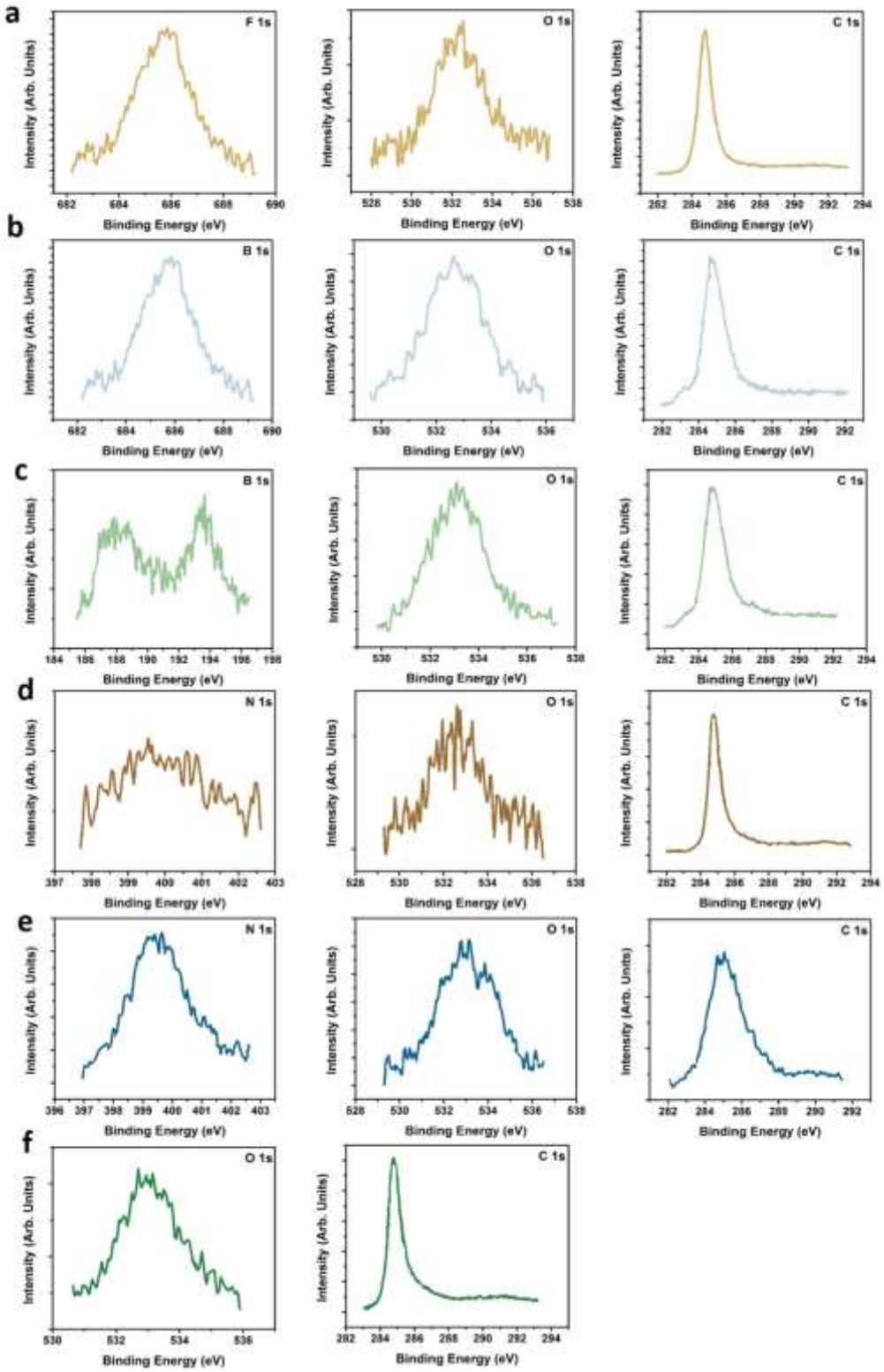

Figure S3. XPS curves for (**a)** F-doped GO, (**b)** pure B B-doped GO, (**c)** C_4B B-doped GO, (**d)** NH_3 N-doped GO, (**e)** C_3N_4 N-doped GO, and (**f)** O-doped GO.

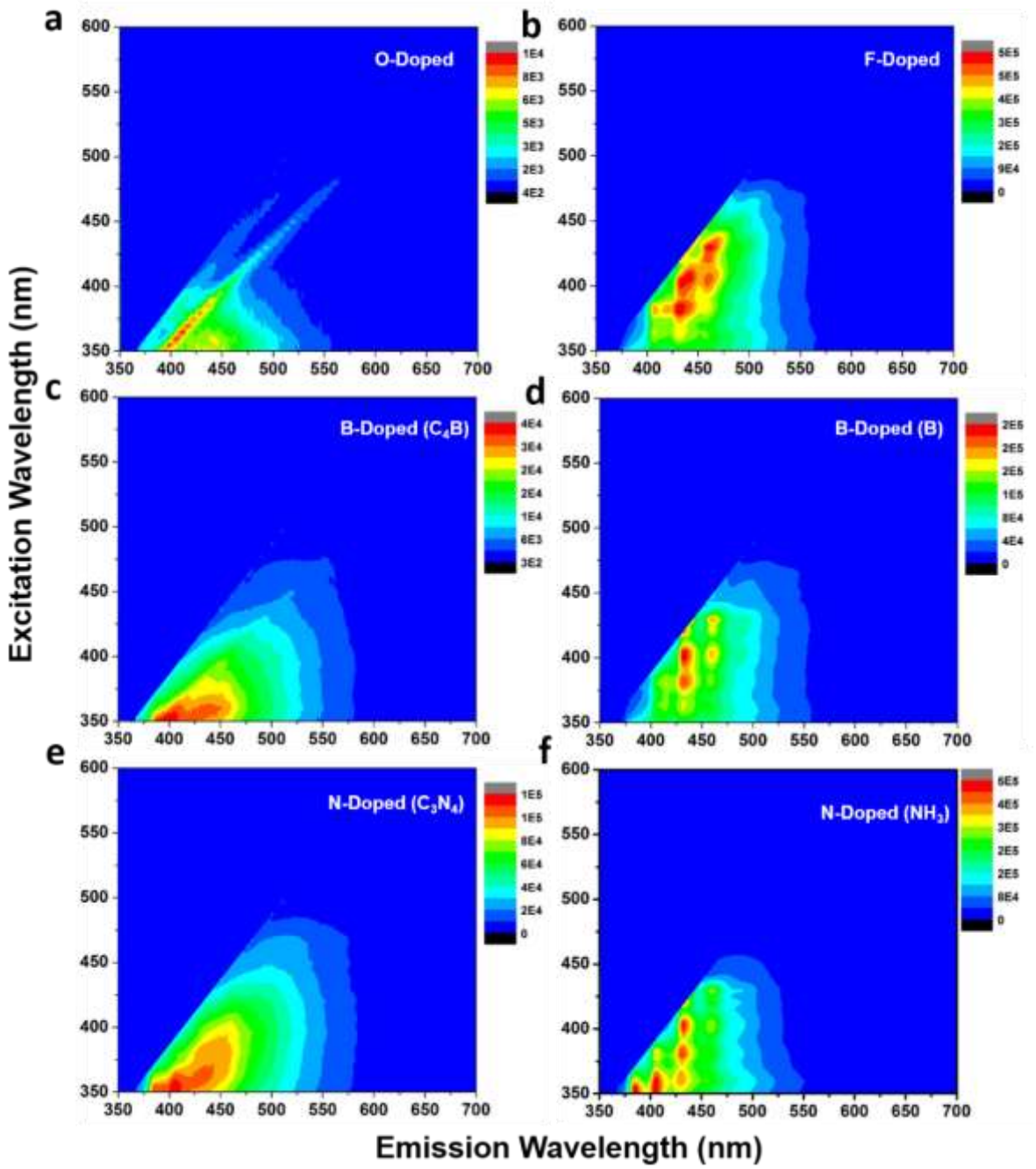

Figure S4. Excitation/Emission curves for the different doped GO samples as marked on each plot. The O-doped sample shows a Raman scattering peak from the solvent.

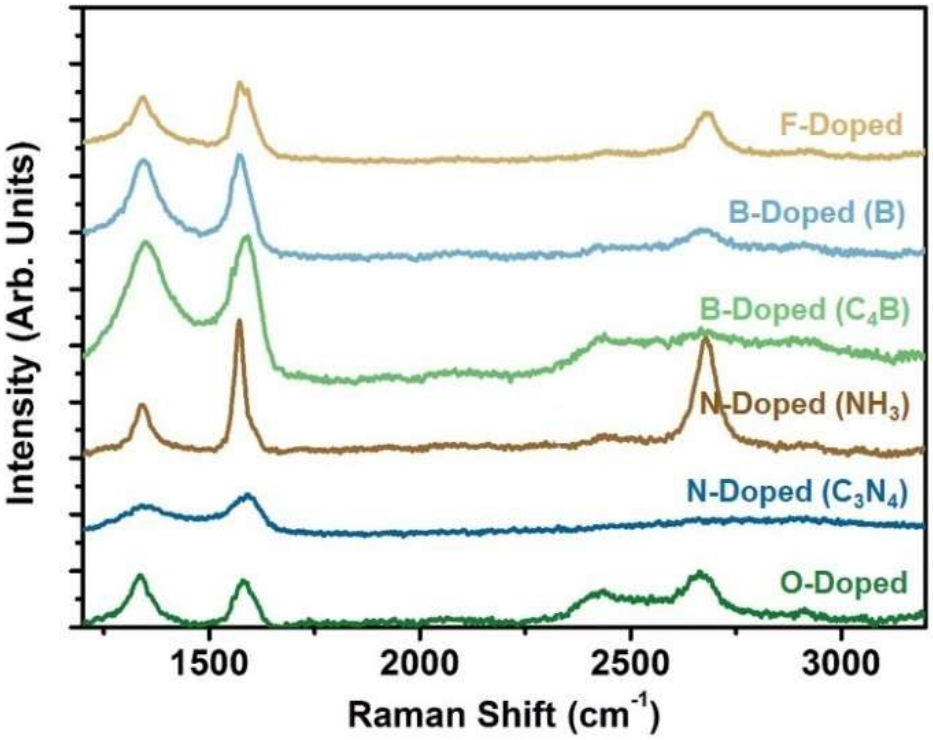

Figure S5. Raman signal for the various doped GO. The peaks at 1345 cm^{-1}, 1570 cm^{-1}, 2450 cm^{-1}, 2680 cm^{-1} and 2910 cm^{-1} are assigned to the D, G, D+D'', 2D and D+D' bands of graphene.

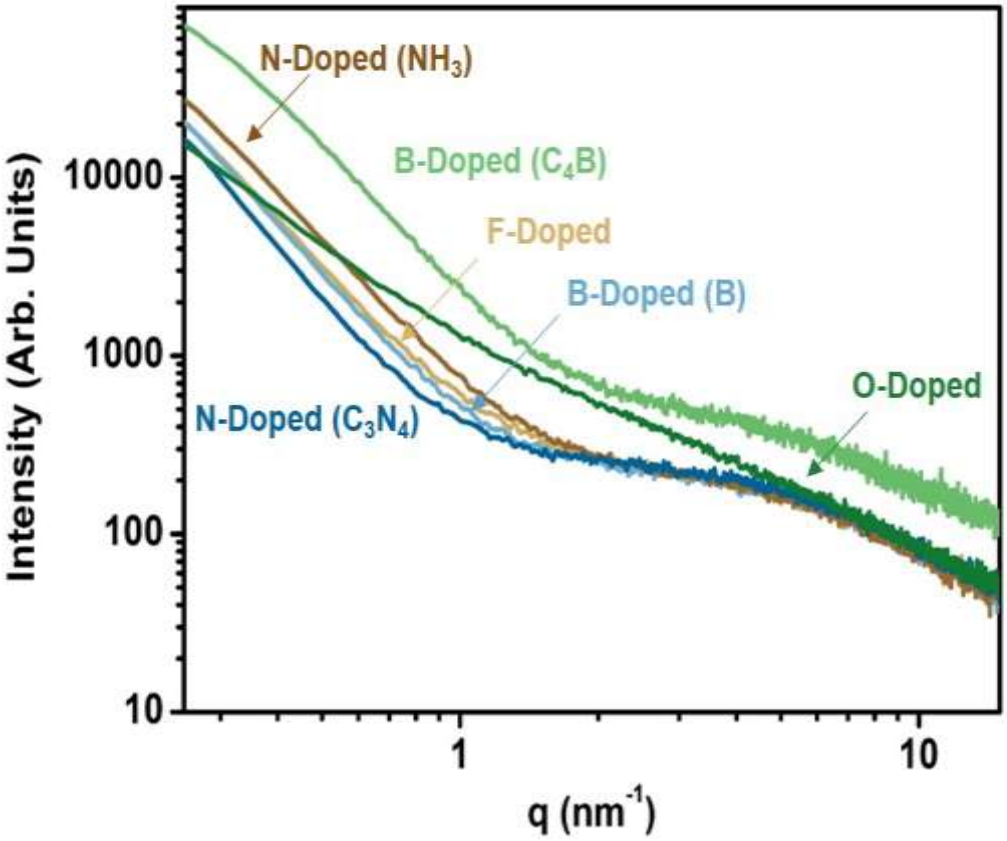

Figure S6. Small-angle X-ray scattering (SAXS) intensity versus the scattering vector (q) for the various doped GO samples.

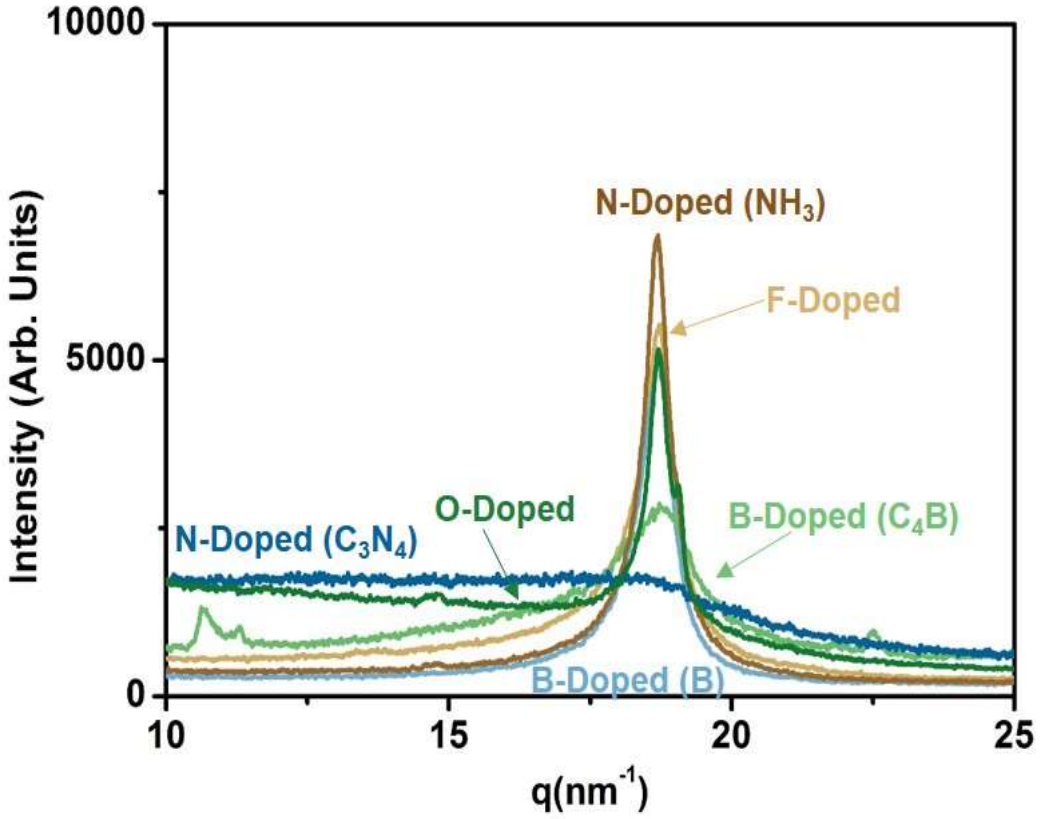

Figure S7. Wide Angle X-ray Scattering (WAXS) intensity versus the scattering vector (q) for the various doped GO samples.

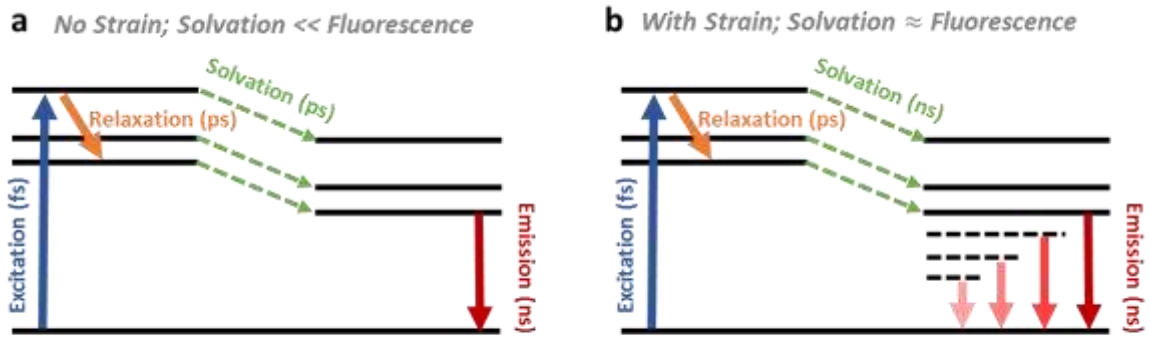

Figure S8. Schematic of fluorescence process without (a) and with (b) strain. Initially, optical transitions occur between the molecular orbitals of functional groups on the GO. These transitions are tunable by changing functional group, defects, and chemical structure. After excitation, carriers quickly thermalize, giving off excess energy within picoseconds. If strain is not present, the external solvent dipoles re-align with the excited state dipole in picoseconds before fluorescence begins, red shifting the final emission wavelength. If strain is present, the excited state dipole and surrounding functional groups on the GO also solvate. This interaction slows solvation to the same time scale as fluorescence, and the emission energy continously redshifts during fluorescence.

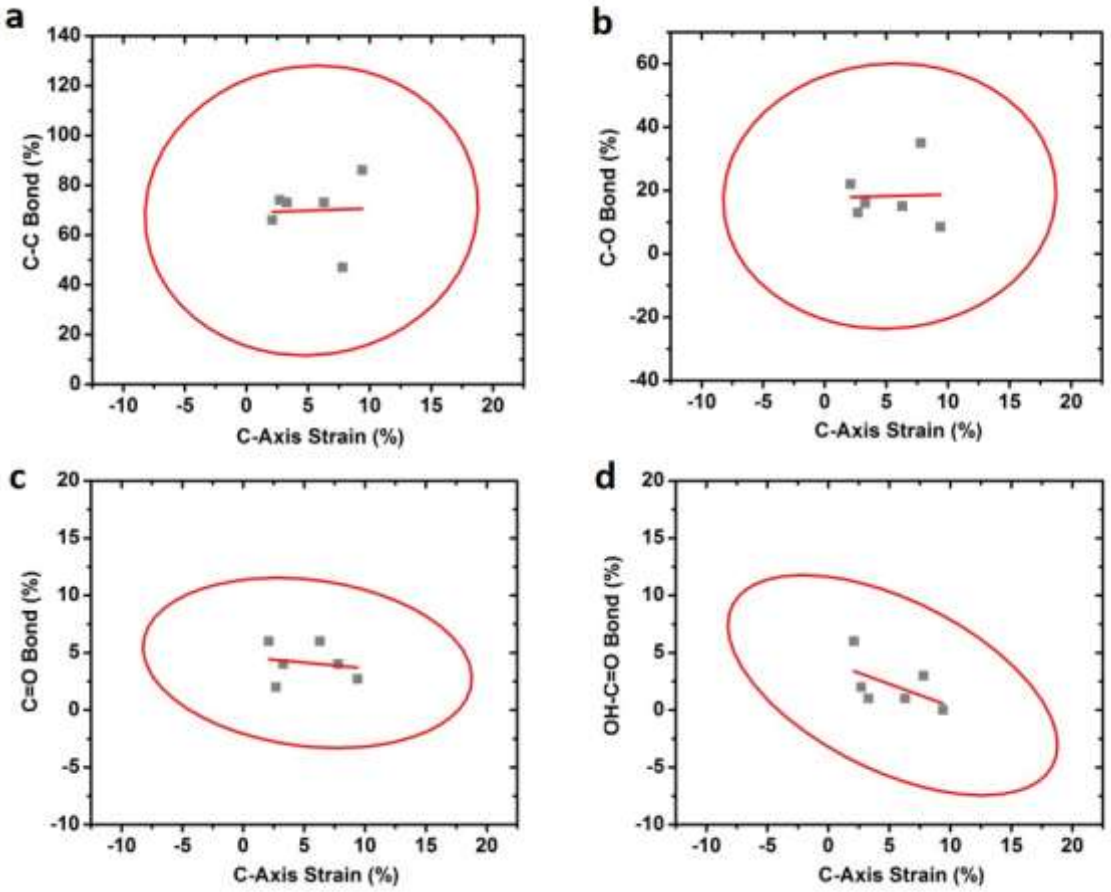

Figure S9. Comparison of correlation for **a,** C-C, **b,** C-O, **c,** C=O, and **d,** -COOH bonding percentage versus the C-axis strain percent, which itself is correlated to the red-edge effect. The ellipse shows the 95% confidence level. The correlation in all cases is less than 0.5. The bonding percentages are shown in Table S4.

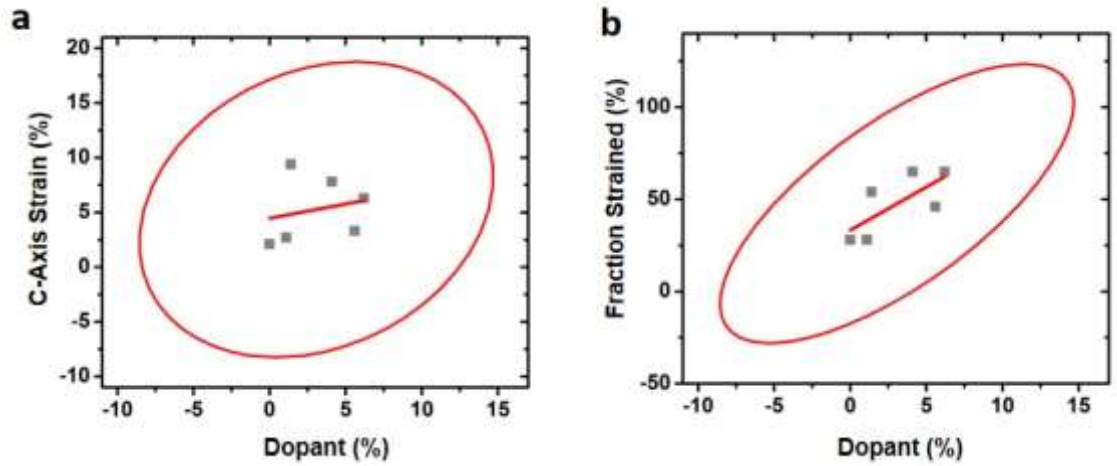

Figure S10. Comparison of correlation for dopant percentage to the **a,** C-axis strain and **b,** the fraction of the GO sheet that is strained. The ellipse shows the 95% confidence level. The correlation in all cases is less than 0.5.

Table S1. Elemental composition of graphene oxide samples

Samples	C [%]	O [%]	F / B / N [%]
F-Doped	95.7	2.9	1.4
B-Doped (B)	87.0	7.4	5.6
B-Doped (C_4B)	85.2	8.6	6.2
N-Doped (NH_3)	97.5	1.4	1.1
N-Doped (C_3N_4)	88.7	7.2	4.1
O-Doped	97.8	2.2	--

Table S2. Raman Information for Id/Ig calculations

Samples	C [%]	O [%]	G Shift from 1585 [cm^{-1}]	2D (G') Shift from 2680 [cm^{-1}]	I_D/I_G	FWHM of D Peak [cm^{-1}]	FWHM of 2D peak [cm^{-1}]
F-Doped	95.7	2.9	-10	-5	0.95	98	81
B-Doped (B)	87.0	7.4	-16	3	1.40	130	201
B-Doped (C_4B)	85.2	8.6	-6	-4	1.74	137	257
N-Doped (NH_3)	97.5	1.4	-15	-4	0.34	43	63
N-Doped (C_3N_4)	88.7	7.2	-1	-7	1.93	175	293
O-Doped	97.8	2.2	-17	-9	0.52	43	63

Table S3. Fraction and Percent Strained

Samples	Fraction of Strained Units [%]	Extension (Strain) Along C Axis [%]
F-Doped	54	9.4
B-Doped (B)	46	2.2
B-Doped (C_4B)	65	6.3
N-Doped (NH_3)	28	2.7
N-Doped (C_3N_4)	65	7.8
O-Doped	28	2.1

Table S4. Bonding Percentage (%) from XPS Analysis

F-Doped	C-C	C-O	C=O	C-F	F-C semi-ionic	F-C covalent	O=C	O-C	O-H
Binding Energy (eV)	284.8	286.0	287.6	290.8	685.5	686.9	531.2	532.2	533.2
Ratio	86.1%	8.5%	2.7%	2.7%	74%	26%	17%	60%	23%

B-Doped (B)	C-B	C-C	C-O	C=O	OH-C=O	B4C	B-O or BC_3	BC_2O or BCO_2	O=C	O-C	O-H
Binding Energy (eV)	283.6	284.8	285.8	287.4	289.3	187.7	189.4	192.9	531.4	532.4	533.4
Atomic Percentage	6%	73%	16%	4%	1%	34%	21%	45%	13%	46%	41%

B-Doped (C_4B)	C-B	C-C	C-O	C=O	OH-C=O	B4C	B-O or BC_3	BC_2O or BCO_2	O=C	O-C	O-H
Binding Energy (eV)	283.6	284.8	285.8	287.2	288.7	187.6	189.4	193.3	532.0	533.0	534.0
Atomic Percentage	6%	73%	15%	6%	1%	37%	24%	39%	16%	49%	35%

N-Doped (NH_3)	C-C	C-O	C-N	C=O	OH-C=O	Pyridinc N	Pyrrolic N	O=C	O-C	O-H
Binding Energy (eV)	284.8	285.7	286.7	287.8	289.5	398.4	399.5	400.8	531.8	532.8
Atomic Percentage	74%	13%	6%	2%	2%	23%	47%	30%	12%	61%

N-Doped (C_3N_4)	C-C	C-O	C-N	C=O	OH-C=O	Pyridinc N	Pyrrolic N	O=C	O-C	O-H
Binding Energy (eV)	284.8	285.6	286.6	287.4	289.9	398.8	399.8	400.8	532.0	533.0
Atomic Percentage	47%	35%	11%	4%	3%	35%	52%	13%	7%	62%

O-Doped	C-C	C-O	C=O	OH-C=O	O=C	O-C	O-H
Binding Energy (eV)	284.8	286.0	288.0	290.8	531.8	532.8	533.8
Atomic Percentage	66%	22%	6%	6%	14%	51%	35%