# **Supplementary Information**

# Plasma Modification of the Electronic and Magnetic Properties of Vertically Aligned bi-/tri- layered Graphene Nanoflakes

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## S1: C K-edge shift in XANES spectra of all FLGs.

The peak positions for graphone is similar to that of pristine FLG, but the absorption edges of nitrophene and siliphene are shifted towards higher and lower energy level that can be seen clearly from the first order derivatives of all the C *K*-edge XANES spectra as shown in figure S1, given below.



Figure S1: C K-edge XANES derivatives of pristine and plasma treated FLGs

#### S2: M-H loop of FLG:Si (Siliphene) without the subtracted diamagnetic background.

In FLG:Si (siliphene), the spectral feature is different than graphone/ N-graphene and is considered as mixed dia- and ferro-magnetic in nature as shown in S2 (given below). It is noted that the diamagnetism here shows uo only because the siliphene is intrinsic; whereas the ferromagnetic is very weak. However, due to presence of mixed dia- and ferro-magnetic nature, we have subtracted the diamagnetic background revealing that the saturation magnetic moment is  $\sim 0.11 \times 10^{-4}$  emu/gm only, significantly lower as compared to graphone/N-graphene and even lower than pure FLG (Ms = 3.47 emu/gm) as shown in the main text in figure 5(d).



**Figure S2:** Magnetic hysteresis loop of FLG:Si (Siliphene) with dia-magnetic background at 300 K and 40 K, respectively.

### S2: Wide Energy Survey Spectra (WESS) of the pristine FLGs

The XPS wide energy survey spectrum (WESS) of all the samples did not show any measureable contaminants, thereby confirming the non-existence of any catalyst material which can possibly contribute to the magnetism observed for these materials. In the WESS shown S3, the elemental quantification results revealed C (98.82±0.30); N (0.69±0.22) and O (0.49±0.30) only. No other elements could be detected in the spectra.



**Fig. S3:** XPS - Wide Energy Survey Spectra (WESS) of the pristine FLGs showing the purity of the samples showing only carbon, nitrogen and oxygen elements.