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## Graphene and graphene oxide nanostructure material for spintronic applications

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We report an investigation into the magnetic properties of hydrogen, nitrogen silicon functionalized vertically aligned few layers graphene (FLG) synthesized by microwave plasma enhanced chemical vapor deposition. The FLGs are hydrogenated at different substrate temperatures to alter the degree of hydrogenation and their depth profile. Nitrogenation and silicon functionalization are occurs using nitrogen plasma and silane gas respectively. The unique morphology of the structure gives rise to a unique geometry in which graphene layers are very different from other widely studied structures such as one-dimensional nanoribbons. Saturation magnetization ( $M_s$ ) of pristine FLG is enhanced by over four (4) and thirty-four (34) times to  $13.94 \times 10^{-4}$  and  $118.62 \times 10^{-4}$  emu g<sup>-1</sup>, respectively, as compared to pristine FLGs ( $M_s$  of  $3.47 \times 10^{-4}$  emu g<sup>-1</sup>), via plasma-based-hydrogenation (known as graphone) and nitrogenation (known as N-graphene) reactions, respectively. However, upon organo-silane treatment on FLG (known as siliphene), the saturation magnetization is reduced by over thirty (30) times to  $0.11 \times 10^{-4}$  emu g<sup>-1</sup>, as compared to pristine FLG. Synchrotron based X-ray absorption near edge structure spectroscopy measurements have been carried out to investigate the electronic structure and the underlying mechanism responsible for the variation of magnetic properties. For graphone, the free spin available via the conversion of the sp<sup>2</sup> / sp<sup>3</sup> hybridized structure and the possibility of unpaired electrons from induced defects are the likely mechanism for ferromagnetic ordering. During nitrogenation, the Fermi level of FLGs is shifted upwards due to the formation of a graphitic like extra -electron that makes the structure electron-rich, thereby, enhancing the magnetic coupling between magnetic moments. On the other hand, during the formation of siliphene, substitution of the C-atom in FLG by a Si-atom occurs and relaxes out the graphene plane to form Si-C tetrahedral sp<sup>3</sup>-bonding with a non-magnetic atomic arrangement showing no spin polarization phenomena and thereby reducing the magnetization. Thus, plasma functionalization offers a simple yet facile route to control the magnetic properties of the graphene systems and has potential implications for spintronic applications.

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