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One-Color Photo-Oxidation and Reduction of Graphene for Rewritable Chemical Patterning

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Chemical patterning on graphene offers a powerful tool for tailoring chemical, optical and electronic natures of graphene, which allows for devices engineered for specific demands. A facile and clean method for oxidation of graphene is reported by Mitoma *et al.,* where a focusing visible laser on graphene under water presence results in oxidation of graphene in a site-specific manner [1]. In this study, we report the photo-oxidation under water and further photo-reduction are achieved with a one-color laser by simply adjusting the laser intensity, and demonstrate rewritable chemical patterning on a graphene canvas.

Graphene monolayer deposited on a glass substrate was covered with pure water (Fig. 1a). A visible laser (473 nm, 5 MW/cm^2) was focused at the water/graphene interface, leading to oxidation of graphene with the diffraction limited spatial resolution (+/-300 nm). We further demonstrated that the oxidized spots could be reduced by focusing the same laser but higher intensity (14.3 MW/cm^2). The site-specific photo-oxidation/reduction could be repeated several times and switched each other by simply adjusting the one-color laser intensity. With laser direct writing technique, we conducted photo-writing a chemical pattern "H" on a graphene canvas (Fig. 1b left). Unlike conventional sample annealing, the photo-reduction allows to remove chemical functionalization at an arbitrary position (photo-erasing, Fig.1 b middle). Thanks to the reversibility of the photo-oxidation/reduction cycle, a new pattern "K" could be written on the site (Fig. 1b right). This study will provide a venue for site-specifically tuning of physicochemical properties of graphene monolayer, allowing flexible design of chemical pattern on graphene monolayer.

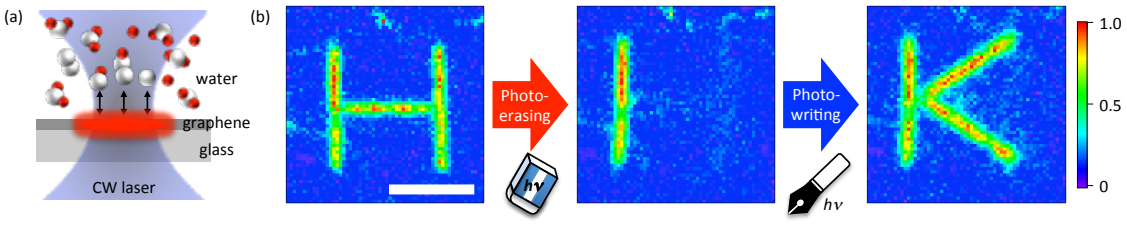

Figure 1. (a) A schematic illustration of photo-oxidation and reduction. (b) A series of Raman I_D/I_G ratio maps, obtained after photo-writing of "H" pattern (left), partially photo-erasing (middle) and sequential photo-writing of "K" pattern on the same site (right). Scale bar 2.5 μm.

[1] N. Mitoma R. Nouchi, K. Tanigaki, *J. Phys. Chem. C,* **2013,** *117*, 1453.