## Ultrafast Time-Resolved Electron Diffraction Capturing Layer Stacking Dynamics of Graphene Oxide

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We demonstrated ultrafast time-resolved electron diffraction measurements on a nanosheet liquid crystal, graphene oxide (GO), to understand their layer stacking dynamics.

Layer stacking of two-dimensional nanosheet materials, *e.g.*, graphene and transition metal dichalcogenides, is critical in controlling their physical and transport properties. GO also undergoes layer stacking along with its photo- and thermal-induced reduction process; however, the mechanism and dynamics during the layer stacking have not been clarified yet. In previous study, we have reported the ultraviolet (UV) photo-induced reduction mechanism (removal of epoxy-group oxygen) of GO using ultrafast time-resolved electron diffraction measurements [1]. In the present study, we demonstrated ultrafast time-resolved electron diffraction to monitor the structural dynamics during the layer stacking of GO induced by UV photoexcitation. The experimental results are combined with the density functional theory calculations, which reveals that AB-stacking of graphitic domains of GO layers coincides in ~40 ps on photo-induced removal of the epoxy-group oxygen from the basal plane of GO *via* overlapping of the  $\pi$ -electronic orbits of GO layers (Fig. 1).



Fig. 1 Electron diffraction pattern from GO thin film (left) and the schematic illustration of the photo-induced layer stacking of GO (right).

## References

[1] M. Hada, et. al., ACS Nano 13 (2019) 10103–10112.