## In-situ Characterisation of Graphene Using Combined XPS and Raman Spectroscopy: Removal of Polymer Residue by Ar Gas Cluster Ion Beams

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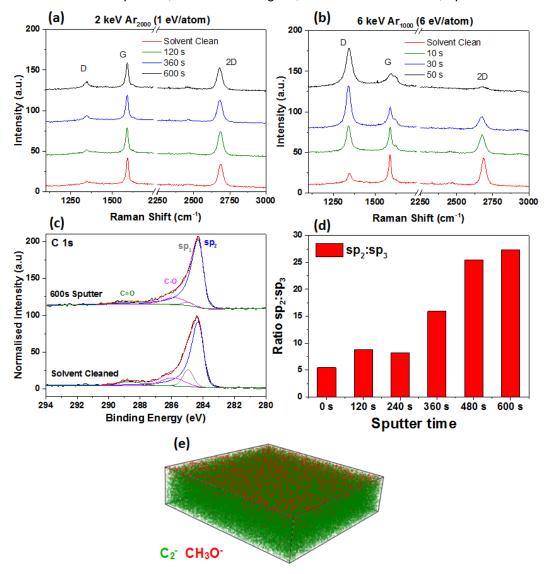


Figure 1. Confocal Raman spectra acquired during sputtering of a graphene surface using (a) 2 keV  $Ar_{2000}$  gas cluster ion beam (GCIB, 1 eV/atom) and (b) 6 keV  $Ar_{1000}$  GCIB (6 eV/atom). While the Raman spectra in (a) show virtually no change during sputtering, the spectra in (b) show a large increase in the D peak, with a concomitant decrease in the G and 2D peaks indicative of the generation of defects in the graphene layer. Peak fitted C 1s XPS spectra acquired concurrently with the Raman measurements from (a) are shown in (c), with the change in sp<sub>2</sub>:sp<sub>3</sub> ratio from these fits shown in (d). These indicate the decrease in the sp<sub>3</sub> carbon peak as the surface of the graphene in cleaned with the argon GCIB.

3D ToF-SIMS imaging of the cleaning process over an imaged area of 150  $\mu$ m × 150  $\mu$ m is shown in (e), again using a 1 eV/atom GCIB on the same sample used for confocal XPS and Raman measurements in (a) and (c). This shows the removal of polymer material from the sample surface as indicated by the CH<sub>3</sub>O<sup>-</sup> (red) signal, while the monolayer graphene C<sub>2</sub><sup>-</sup> (green) signal is observed to remain over the same sputter period.