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Electrical conductivity, chemistry and bonding alternations under graphene oxide to graphene transition as revealed by in-situ TEM

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Summary

A specially designed suspended graphene oxide (GO) device has been fabricated and investigated in a transmission electron microscope (TEM)-scanning tunneling microscopy (STM) based setup. The detailed study of step-by-step slow reduction of an individual GO sheet under current flow and thereby Joule heating, while in-parallel performing conductivity measurements, atomic structure imaging, chemical composition, and bonding alternations tracing have been performed. As monitored by electron energy core loss spectroscopy, the oxygen content is tuned from that peculiar to a pristine GO, i.e. 23.8 at.%, to oxygen-free pure graphene. Six orders of magnitude conductance rise is achieved during this process with the final conductivity reaching 1.5A-10$\hat{}${}5 S/m. Quantification of plasma energy losses of the starting GO shows that
{}{}40% of the oxygen atoms are in the form of epoxy, while
{}{}60% oxygen atoms are in the form of hydroxyl, and the total portion of sp3 bonds in pristine GO is estimated to be
{}{}45%. The epoxy groups show a larger influence on the conductivity of GO than hydroxyl ones. From the plasma-loss spectra oxygen atoms in epoxy groups of GO are found to decompose before those in hydroxyl groups under a phase transformation to graphene.