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Whelan, Patrick Rebsdorf; Kostesha, Natalie; Larsen, Martin Benjamin Barbour Spanget; Balogh, Zoltan Imre; Bøggild, Peter; Booth, Tim

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Characterization of copper oxidation at graphene/copper interfaces by transmission electron microscopy

Patrick R. Whelan\textsuperscript{1}, Natalie Kostesha\textsuperscript{1}, Martin B. Larsen\textsuperscript{1}, Zoltan I. Balogh\textsuperscript{2}, Peter Bøggild\textsuperscript{1}, Timothy J. Booth\textsuperscript{1}

\textsuperscript{1} DTU Nanotech, Technical University of Denmark
Ørsteds Plads, Building 345E, 2800 Kgs. Lyngby, Denmark
patwhe@nanotech.dtu.dk

\textsuperscript{2} DTU Cen, Technical University of Denmark
Fysikvej, Building 307 and 314, 2800 Kgs. Lyngby, Denmark

High quality, large-area graphene can be grown on the surface of Cu catalyst layers by chemical vapor deposition (CVD) \cite{1}. Despite graphene being impermeable to gases \cite{2}, the Cu catalyst layer beneath an incomplete or cracked CVD grown graphene sheet will oxidize when exposed to ambient conditions \cite{3,4}.

In this study we use a focused ion beam to lift-out lamellae of graphene grown by CVD on the Cu(111) surface, and find two distinct copper oxide structures, depending on whether the oxide lies beneath graphene or on an area of exposed Cu surface. We find Cu$_2$O layers oriented parallel to the Cu(111) surface beneath graphene which are typically 1.5 - 2 nm thick with an interlayer distance corresponding to the Cu$_2$O lattice constant. Cu$_2$O on the Cu(111) surface without graphene resembles a Cu$_2$O(111) structure \cite{5} with a thickness of 3 nm.

Based on these results we present a transfer technique which proceeds by oxidation of the copper catalyst layer beneath the graphene, decoupling the graphene from the catalyst surface by reducing the binding energy. The mechanism of formation of oxide layers beneath single atomic thick passivation layers such as graphene is also important for the potential use of graphene and other two dimensional materials as anti-corrosion or bio-compatible coatings.

References:

\cite{1} X. Li et al., Science, 324 (2009) p. 1312.
\cite{2} J. S. Bunch et al., Nano Letters, 8 (2008) p. 2458.
\cite{3} P. Kidambi et al., Nano Letters, 13 (2013) p. 4769.
\cite{4} A.-Y. Lu et al., RSC Advances, 2 (2012) p. 3008.

Figure 1. Top left shows a SEM image of graphene grown on a Cu/SiO$_2$/Si substrate. Most of the sample is covered by graphene (blue arrows), while there are some areas where the graphene has not grown together (yellow arrows). The red rectangle, which contains areas with and without graphene and a grain boundary, shows an area where a lamella has been cut out. Bottom left and right are TEM images of a Pt/graphene/Cu/SiO$_2$/Si lamella. Bottom left shows a Cu grain boundary. Top right shows an area where Cu is covered by graphene, while bottom right shows an area with no graphene. Insets show zoom in on the Cu oxide area and the distance between layers. It is seen that there is a difference in the Cu oxide structure for Cu with and without graphene.