

***In Situ* Characterisation of Graphene Growth on Liquid Metal Catalysts by Chemical Vapour Deposition**

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Two-dimensional materials (2DMs) with their benchmark – graphene – present one of the most studied material families and have bright perspectives for advanced applications in many technological fields. However, the unique properties of 2DMs largely depend on their quality, that is, the absence of defects and the size of individual crystals. The current state-of-the-art synthesis method of 2DMs is based on chemical vapour deposition (CVD) on a solid catalyst. This process is slow by nature, inefficient, and environmentally unfriendly. The produced 2DMs are often defective, contaminated, and suffering from very high residual stress, compromising their unique physio-chemical properties for the expected applications. Liquid metal catalysts (LMCats) such as molten Cu thanks to their atomically smooth surface and fast mass transport help to overcome these limitations and allow the preparation of large-area single-layer graphene. Furthermore, graphene flakes show the tendency to self-align on a liquid substrate. The study of such systems, however, typically required cooling down to room temperature and the re-solidification of the samples, altering the surface significantly. In this way, the information on the dynamic of the growth was lost entirely.

In a collaboration between several scientific teams across Europe, a customized mobile CVD reactor has been developed and adapted to harsh experimental conditions such as high temperature (1400 K) and metal sublimation rate for *in situ* characterisation combining X-ray Reflectivity (XRR) and other X-ray scattering techniques, Raman spectroscopy, and radiation mode optical microscopy [1,2]. As a result, we demonstrate that the graphene growth can be followed and tailored in real time with high reproducibility. By varying the growth conditions in a broad range of temperatures and gas partial pressures, we examine the growth kinetics of graphene domains on liquid Cu. Thanks to the synchrotron beam and a new numerical method for analysis of XRR on curved surfaces [3], we are able to accurately characterise the physicochemical properties of the graphene layer at the atomic level such as its adsorption height above liquid Cu [4]. We also explore the growth of graphene on low-melting-temperature CuGa alloys as an alternative to Cu. The gained knowledge is essential for developing efficient mass production and transfer technologies of 2DMs using the LMCats.

References

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