Synthesis Of High-Quality Graphene Using Boudouard Reaction

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The breakthrough properties of single-layer graphene stimulated the increased interest of the scientific community and industry in scalable graphene production and wide application of devices based on it. In particular, graphene is a promising material in optoelectronics, photovoltaics, membrane technologies, catalysis and electron microscopy. At the same time, the unique characteristics of graphene (mobility of charge carriers, conductivity, mechanical strength) are limited by defects and, in particular, grain boundaries. This stimulates the need for development of methods for defect-free single-crystal graphene. Though the mechanical exfoliation of highly oriented pyrolytic graphite allows to obtain graphene of the best quality, it has limited scalability. Therefore, chemical vapor deposition on the catalyst surface (CCVD) is recognized to be the most promising approach to the scalable synthesis of single-layer single-crystal graphene. At the same time, stable carbon sources (usually methane) and metals with low carbon solubility (for example, based on copper) are usually employed to suppress the formation of the second layer.

In this work [10.1002/advs.202200217], we use a combination of physicochemical methods (scanning electron microscopy, atomic force microscopy, terahertz and optical spectroscopy, Raman spectroscopy, X-ray photoelectron spectroscopy, reflected electron diffraction, slow electron diffraction, *etc.*), the features of nucleation and growth were studied graphene crystals on a copper catalyst, and the structure of the catalyst surface was also revealed. The kinetic data obtained in combination with modeling based on the DFT method allowed us to highlight the rate-limiting processes and propose new models of graphene nucleation and growth. Moreover, fine-tuning of graphene production conditions (temperature, composition of the reaction medium, pressure in the reactor) opened the way to the original technology for producing exclusively single-layer graphene with centimeter-sized crystallites (Figure 1).



Figure. 1. Optical micrography (a) and photography (b) of graphene single crystals on the Cu surface.

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