

## Controlling the Nucleation Site of Single-Crystal Graphene on Cu foil Sandwiched by Ni

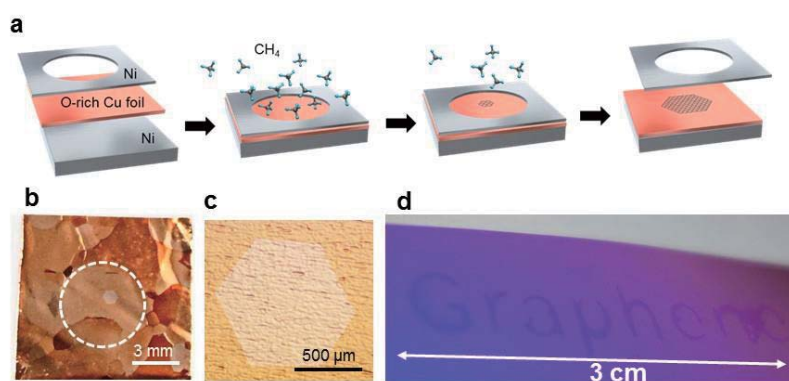
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Graphene grown on Cu by chemical vapor deposition (CVD) is one of the most promising approaches to produce large-area graphene at relatively low cost. However, CVD-grown graphene is composed of merged small grains that are separated by grain boundaries (GBs), which degrade the physical and electrical properties of graphene.<sup>1,2</sup> Controlling the position of the nucleation allows to obtain single graphene grains without GBs. However, it is still a challenge to precisely control the nucleation sites on Cu foil. In addition, the graphene grown on Cu foil usually contains many areas of bi-/few-layer graphene, which further decreases the quality of graphene.<sup>3</sup>

Here, we demonstrate that the nucleation site of graphene on Cu can be effectively predetermined by placing a perforated Ni foil mask on the top during the CVD (Figure 1a), used to control the local concentration of CH<sub>4</sub>.<sup>4</sup> This configuration allows to grow an isolated millimeter-sized single-crystal graphene grain at the designated spot on the Cu surface (Figure 1b,c). The formation of bi-/few-layer graphene areas can be completely suppressed at the same time, by placing a second Ni foil underneath the Cu. Our method can be extended to obtain arrays of large isolated graphene grains by employing multi-holed Ni masks. By prolonging the CVD time we also found that graphene can extend its growth to the edges of Ni, resulting in the direct growth of patterned graphene (Figure 1d). To prove the validity of this method, an array of graphene field-effect transistors (FETs) was successfully fabricated on a 4-inch Cu film without using any lithography processes. Our strategy offers a facile and low-cost way for controlling the nucleation sites of graphene on Cu, promoting the industrialization of graphene in the future.



**Figure 1.** (a) Illustration of the strategy for the spatially controlled growth of single-crystal graphene grains on Cu. The Cu is sandwiched between two Ni foils during the CVD. (b,c) As-grown, mm-sized single graphene grain obtained by using the method shown in (a). (d) Patterned growth of the word “Graphene” by using a corresponding Ni mask.

### References:

[1] Huang, P. Y. et al., *Nature* **469**, 389 (2011). [2] Ago, H. et al., *ACS Nano* **10**, 3233 (2016). [3] Han, Z. et al. *Adv. Funct. Mater.* **24**, 964 (2014). [4] Ding, D. et al., *ACS Nano* **10**, 11196 (2016).