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Scalable growth of single-crystal Graphene on poly-crystalline metal

Abstract
Many studies of graphene have shown that it has enormous potentials for next-generation technology. However, its cost-efficient, reliable, and high-throughput synthesis of high-quality single-crystal graphene is highly required for practical applications of its superb properties. In this paper, we introduce a new seeded growth approach for the scalable growth of high-quality single crystal of graphene on cost-effective polycrystalline metal substrates. In this seeded growth is similar to transplantation of rice seeding, which involves transplantation of small single crystal seeds to large scale polycrystalline catalyst substrate and their subsequent in-plane homo-epitaxial growth to a uniform single crystal graphene layer. This approach is scalable and can be used for multi-meter scale growth of single crystal graphene. In addition, by defect healing process, it is possible to ensure the significantly low density of defects on the 2D material.

1. INTRODUCTION

Graphene, a representative two-dimensional (2D) material, has attracted intense interests due to its extraordinary physical and chemical properties.[1] Unique band structure and electronic properties of graphene make it promising for applications in next-generation electronic devices, transparent flexible conducting electrodes, and sensors. The extraordinary properties and potential applications of graphene have motivated the development of large-scale, synthetic graphene grown by various methods. In particular, it has been shown that polycrystalline and predominantly monolayer graphene can be catalytically grown by CVD in large scale.[2] However, non-uniform distribution of defects in poly-crystalline graphene significantly affect reliability of its devices, and thus high quality single-crystal graphene is strongly required for the high-end industrial applications such as high-performance electronic devices. A typical approach for the growth of single crystal graphene involves growing a single grain to as large a size as possible from a single nucleation site through inhibition of different graphene nucleation.[3] However, this approach is not suitable for reproducible and high-throughput synthesis. The second approach is catalytic and epitaxial growth of graphene on a single crystal substrate. Alternatively, wafer-scale single-crystal graphene has been epitaxially grown on single crystal germanium substrate.[4] However, this method also has practical drawbacks, such as size limit and cost-demanding process. Herein we introduce “seed transplantation” approach for the growth of a single crystalline graphene layer on polycrystalline metal substrate.

2. RESULTS AND DISCUSSION

Overall “seed transplantation” process for the growth of single crystal graphene was shown in Figure 1. At first, single crystal graphene seeds are grown on single-crystal Ge surface using previous reported method.[4] Single crystal graphene seeds were then selectively transferred to polycrystalline substrate by μ-contact printing. For reliable contact printing of the seeds, we deposited Au on the graphene seeds and also used polydimethylsiloxane (PDMS) mold treated with self-assembled monolayer. Thus Au/graphene seeds were selectively detached by designed PDMS mold[5] and transferred to the polycrystalline Pt-foil (3×3 cm2) by hot press printing. The two processes were repeated for transplanting on large substrate. After the seed contact printing, transferred seeds homo-epitaxially grew without further random nucleation to a uniform single-crystal graphene layer using controlled CVD process. In the CVD process, carbon precursor preferred to diffuse to seed edge that was the starting point for the growth because the energy required for edge growth is lower than required to graphene nucleation energy.[6] By inducing in-plane homo-epitaxial growth of the graphene seeds and inhibiting additional nucleation of graphene, which may have different crystal orientation,[7] graphene seeds were expanded and merged to form a large area single-crystal graphene.

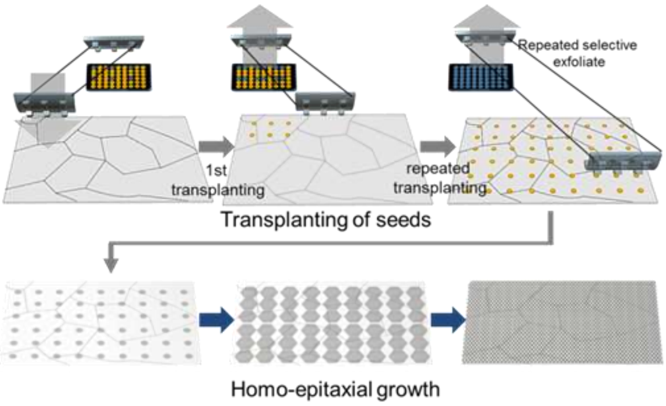

Figure 1: Schematic illustration of growth process of single crystal graphene on polycrystalline metal surface, which involves (a) Seed contact printing and (b) subsequent in-plane homo-epitaxial growth. In process of seed contact printing

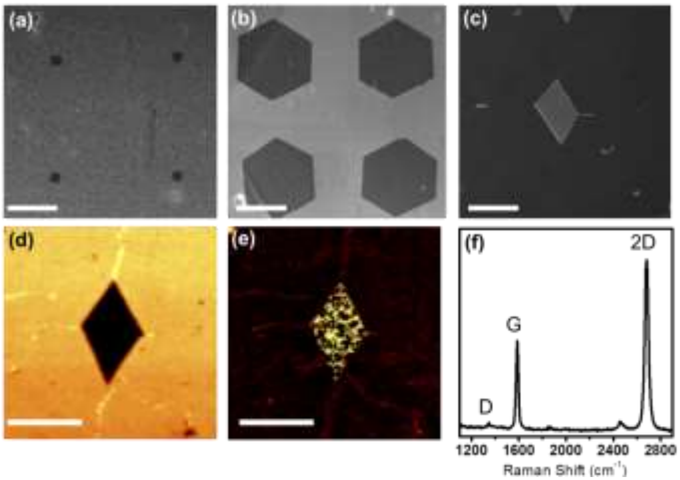

Figure 2: SEM images of seeded growth process of graphene. (a) transferred seed graphene on metal foil, (b) homo-epitaxial growth graphene, (c) merging graphene seeds. Raman spectrum analysis of merging graphene, (d) mapping image of 2D peak (e) D/G ratio and (f) Raman spectrum at the merging area. (Scale bar (a), (b), (c): 50 μm, (d), (e): 40μm)

Figure 2(a) shows SEM image showing that the single graphene seeds were safely transferred to Pt foil. And through homo-epitaxial growth, Figures 2 (b) and (c) demonstrate that the graphene seeds grew homo-epitaxially to uniaxially aligned single-crystal domains without additional nucleation and then the domains were subsequently merged also when they merged. Raman mapping images of 2D (Fig. 2 (d)) and D/G (Fig. 2 (e)) peaks clearly indicates the uniaxially grown graphene domains were merged to single crystal graphene without grain boundary defects.

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