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PERSPECTIVE

Growth and applications of graphene on copper

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Since the first isolation of graphene by Geim and Novoselov in 2004, this two-dimensional (2D) material has attracted significant attention from the scientific and engineering communities.[1] Due to its structure with a single layer of covalently bonded sp^2-hybridized carbon atoms, graphene has superior mechanical, electrical, optical, and thermal properties.[2–4] Although mechanical exfoliation remains an ideal method for obtaining high-quality graphene, its scalability is severely limited; thus, it is impractical for industrial applications. In pursuit of a method suitable for large-scale production, research has been performed using chemical vapor deposition (CVD) methods for synthesizing graphene on metal substrates.[5,6] Among the various metal substrates used for graphene growth, copper (Cu) is the preferred choice because of its small lattice mismatch (2.556 Å for Cu(111) and 2.46 Å for graphene[7]), low carbon solubility (<0.02 at.%[8,9]), and cost-effective characteristics. By investigating the advancements in graphene growth on different types of Cu materials, this article aims to shed light on the current challenges and future directions in the field, thereby serving as a cornerstone for researchers and technologists interested in graphene synthesis and its applications.

In principle, three types of Cu materials have been identified as suitable for graphene growth; these include liquid Cu, Cu films, and Cu foils, and each has unique advantages for graphene synthesis. Liquid Cu is favored for its inherent fluidity and absence of grain boundaries, facilitating a "self-assembly process" conducive to the growth of single-crystal graphene. Solid Cu is more appropriate for large-scale manufacturing, although a single-crystallization process is needed to support the epitaxial growth of single-crystal graphene. Among solid Cu substrates, Cu films are notable for their flatter microscopic surface, which enhances the quality of graphene and simplifies its subsequent transfer process; moreover, Cu foils are readily available in large quantities, and this aspect is crucial for the synthesis of large-scale single-crystal graphene. In summary, with the development of orientation control based on liquid Cu and the fabrication technique of single-crystal solid Cu, a foundation has been established for the growth of large-scale single-crystal graphene, representing a significant achievement in the field.

Liquid Cu has emerged as a promising substrate for fabricating high-quality single-crystal graphene.[10] The synthesis of graphene on liquid Cu necessitates the initial

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melting of solid Cu into a liquid state, eliminating the grain boundaries in the Cu substrate and facilitating uniform nucleation of graphene islands across the substrate. The inherent high fluidity of liquid Cu also allows the graphene islands to rotate and move freely, promoting alignment in the same orientation to form a single-crystal graphene film[10,11] (Figure 1a). Recently, because of the unique properties of liquid Cu, self-aligned graphene nanoribbons with widths down to sub-10 nm and aspect ratios up to 387 were fabricated via a template-free CVD route.[12] In addition, the growth of single-crystal twisted bilayer graphene on liquid Cu was achieved,[13] exhibiting superior thermal stability and carrier mobility as high as 26,640 $cm^2\ V^{-1}\ s^{-1}$. Overall, the dynamic and rheological nature of the liquid Cu enables the graphene islands to rotate, move, and align freely, which provides an important platform for the orientation control in the growth of single-crystal graphene.

Cu films are suitable for the preparation of high-quality graphene for 2D device applications due to their smooth surfaces and the simplicity of subsequent device fabrication processes. For the growth of graphene on Cu films, the first step is to deposit Cu on insulating substrates through a sputtering process. This process is followed by an essential annealing treatment to produce a single-crystal Cu film with a smoother surface. Then, the carbon atoms can diffuse on the surface, and graphene can nucleate with the same orientation and merge as a single-crystal film following the epitaxial relationship. The obtained graphene film can be fabricated directly on the insulator substrates by etching away the residual Cu film[14] (Figure 1b). Recent advancements have introduced several methods for the fabrication of highly smooth single-crystal Cu films to further improve the quality of graphene. Notably, a Cu(111) film with a mono-atom step-level flat surface and a semi-permanent oxidation-resistant property was fabricated by atomic sputtering epitaxy with a single-crystal Cu(111) target.[15] Another innovative approach involves directly converting polycrystalline Cu foils into single-crystal Cu films with smoother surfaces via a prolonged annealing procedure on c-sapphire.[16] These Cu films of superior quality are particularly suitable for the growth of wrinkle-free and adlayer-free graphene, promoting the transfer of ultra-smooth graphene, and contributing to high-quality electronic devices.[17] With these smooth graphene films, the average carrier mobility of CVD graphene-based field-effect transistors is much greater (~8000 $cm^2\ V^{-1}\ s^{-1}$). In summary, Cu films are instrumental in the wafer-scale production of graphene and play a vital role in the integration of 2D devices.

For graphene synthesis on Cu foils, the availability of large-scale commercial Cu foils presents a significant advantage. The Cu foils' adjustable thickness allows for more adaptable annealing treatments, facilitating the production of large-scale single-crystal Cu foils. Recently, large-scale single-crystal Cu foils with different indices were successfully fabricated through a seeded growth method, providing crucial and various substrates for the growth of single-crystal graphene.[18] By utilizing these single-crystal Cu foils as substrates, graphene can nucleate and grow with the same orientation and finally merge into a single-crystal film (Figure 1c). The increased availability of Cu foils has prompted considerable

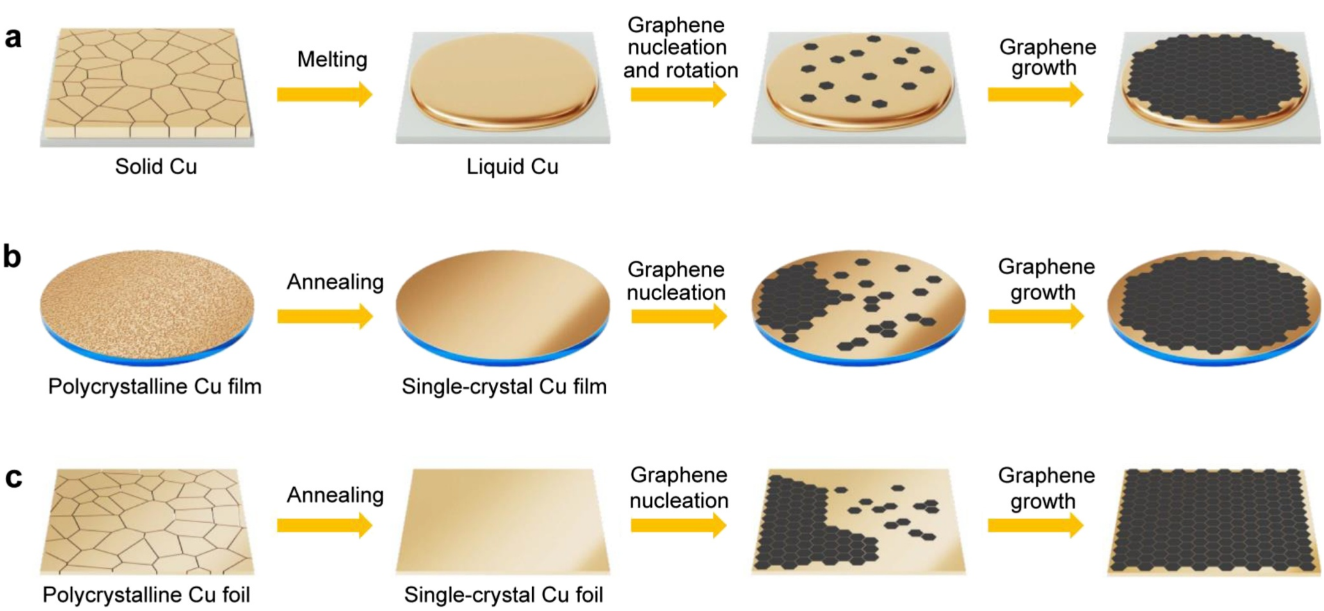

FIGURE 1 Schematic diagrams of graphene growth process on (a) liquid Cu, (b) Cu films, and (c) Cu foils.

research advancements, including the achievement of adlayer-free and wrinkle-free graphene, the achievement of multilayer graphene growth, and the growth of graphene on polycrystalline Cu foils. (1) With a pre-annealing process to remove the subsurface carbon[19] or control the supply of oxygen,[20] an adlayer-free graphene film was successfully obtained on Cu foil. (2) With a lower growth temperature (<1030 K) and a Cu-Ni(111) alloy foil substrate[21] or a proton-assisted method,[22] wrinkle-free graphene films were obtained. (3) Through the introduction of CO_2, the direct growth of continuous bilayer graphene with a high ratio of AB-stacking structures was achieved,[23] and large-scale twisted bilayer graphene with tunable twist angles was successfully fabricated with the assistance of Cu(111) foil.[24] (4) In addition to single-crystal Cu foils, single-crystal graphene could also be synthesized on Cu foils with twin crystals. Through analysis of the geometrical relationships of the twin crystal planes, it is found that the single-crystal graphene could be epitaxially grown on specific twinned substrates.[25,26] In short, the growth of large-scale high-quality graphene films on Cu foils has been achieved. The prospect of producing large-scale, high-quality, single-crystal graphene on Cu foils via a roll-to-roll process appears to be feasible, indicating a new era of scalable and efficient graphene production.

The capability for volume production is a significant advantage of graphene growth on Cu substrates for industrial applications, especially for the integration of graphene into 2D devices in the electronics, optoelectronics, and acoustics fields (Figure 2). Recently, due to

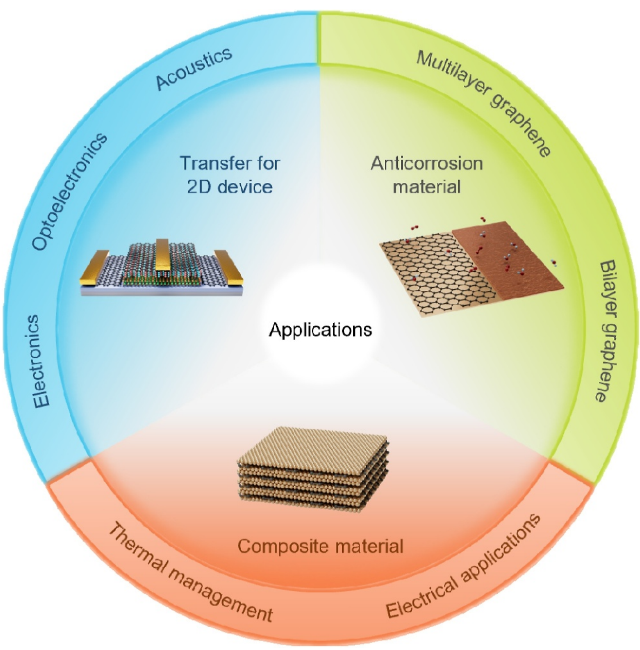

FIGURE 2 Applications of graphene growth on Cu.

the improved quality of graphene grown on Cu, more researchers have opted to use these materials for fabricating 2D devices. By choosing the graphene edge as the gate electrode, sidewall 2D transistors with sub-1 nm gate length were successfully fabricated with an on/off ratio of up to 1.02×10^5.[27] Additionally, using high-quality graphene in a back-gated configuration, hybrid molecular graphene field effect transistors as the optoelectronic platform were also realized.[28] By applying a photonic crystal structure and graphene as a waveguide, a highly efficient modulator was designed with a high modulation depth of 98%.[29] Furthermore, the use of suspended graphene resulted in the fabrication of capacitive acoustic sensors with higher sensitivity and better response.[30] In addition, developing transfer technology is crucial for advancing the applications of graphene films for 2D devices. Techniques such as gradient surface energy modulation[31] or wafer bonding methods[32] now enable the transfer of graphene films with improved quality, facilitating their integration into devices. This evolution demonstrates the growing potential of graphene in the fabrication of high-performance sophisticated 2D devices, highlighting the critical impact of the Cu substrates in enabling the high-volume production necessary for future applications.

Graphene itself also exhibits inherent anti-corrosion properties when coated on metal surfaces. Few-layer graphene has been confirmed to be an effective protective coating layer, primarily because of the misalignment of its grain boundaries, which prevents the vertical infiltration of corrosive molecules onto the Cu surface.[33] A graphene film with no ripples or wrinkles further amplifies these anti-corrosion capabilities by avoiding the "step-induced diffusion" in wet corrosion.[34] However, the anticorrosion efficacy of single-layer graphene remains debatable, with some studies even demonstrating that graphene may promote corrosion in the long term.[35,36] Recent research has revealed the unique Janus doping effect of bilayer graphene, which can protect Cu substrates for more than 5 years at room temperature.[37] In this bilayer graphene/Cu scheme, the bottom layer of graphene is heavily doped and forms a strong interaction with Cu, which limits the interfacial diffusion of oxygen; however, the top layer of graphene is nearly charge neutral, which protects Cu from galvanic corrosion. This innovative approach highlights the potential of graphene coatings to significantly enhance the durability of metal substrates against corrosion, providing opportunities for their encapsulation in the integrated circuits industry. These developments represent a promising avenue for utilizing graphene's unique properties to enhance the performance and durability of electronic components and devices.

Furthermore, graphene, in conjunction with its Cu substrate, can function as an advanced composite material that is particularly suitable for conductive applications.[38] In the field of thermal management, the high degree of alignment of graphene nanosheets with Cu has demonstrated a significantly improved in-plane thermal conductivity (458 W m K^{-1}) and a reduced through-plane coefficient of thermal expansion (6.2 ppm K^{-1}), providing promising prospects for enhancing heat dissipation capabilities.[39] In applications requiring high electrical conductivity, a hot-pressed laminated graphene/Cu composite material has achieved an electrical conductivity of approximately 117% of the International Annealed Cu Standard.[40] Moreover, the innovation of an axially continuous graphene-Cu wire, in which the graphene envelops the surface of the Cu wire, provides superior characteristics such as enhanced surface heat dissipation (224% higher than that of the Cu wire without graphene), increased electrical conductivity (41% higher than that of the Cu wire without graphene), and improved thermal stability.[41] These advancements show the potential of graphene and Cu composites for pushing the boundaries of material performance (especially thermal and electrical conductivity) in industrial applications.

Overall, Cu has been proven to be a suitable substrate for the growth of graphene; this provides a pathway for the scalable and economical production of high-quality graphene films, thus accelerating the industrial application of high-quality graphene in various sectors. In the future, there are four key perspectives for the advancement of graphene growth on Cu. (1) The mechanism of graphene growth and the influence of CVD parameters have already been extensively studied; however, a deeper understanding of the interfacial coupling between graphene and Cu has not yet been fully understood. (2) Despite ongoing research into the use of large-scale graphene/Cu composite materials for conductive applications, the real industrial application of this material continues to be a significant challenge. (3) As the quality of the Cu substrate improves, graphene with fewer defects for transfer to 2D devices can be produced; however, further research is needed to develop cleaner and less damaging methods for transferring graphene and simplifying the fabrication process. (4) By combining the increase in conductivity with the anticorrosion surface of bilayer graphene, graphene/Cu composite materials with superior performance can be expected. In summary, the growth of graphene on Cu facilitates high-end industrial applications of high-quality graphene, with promising opportunities for discovery and advancement.

AUTHOR CONTRIBUTIONS

Chong Zhao: Investigation; writing - original draft; conceptualization. **Meng-Ze Zhao**: Investigation; conceptualization; writing - review & editing. **Zhi-Bin Zhang**: Investigation; writing - review & editing. **En-Ge Wang**: Project administration. **Kai-Hui Liu**: Funding acquisition; project administration. **Mu-Hong Wu**: Funding acquisition; project administration.

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CONFLICT OF INTEREST STATEMENT

Kai-Hui Liu is an editorial board member for cMat and was not involved in the editorial review or the decision to publish this article. The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

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