EDITORIAL

Graphene: Two decades of revolutionizing material science

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The discovery of graphene, a single layer of carbon atoms arranged in a two-dimensional (2D) honeycomb lattice, represents one of the most significant advancements in material science in the 21st century. Although theoretical exploration of graphene has been underway for many years, it was not until 2004 that a major breakthrough occurred. Andre Geim and Konstantin Novoselov (Figure 1) at the University of Manchester isolated graphene using a simple method known as "mechanical exfoliation", which involved peeling layers from bulk graphite using Scotch tape to obtain a monolayer.[1] This seminal discovery, for which they received the Nobel Prize in Physics in 2010, sparked a global surge in research on 2D materials. Subsequently, it led to the discovery of moiré superlattices, which are recognized for their unprecedented controllability. Now, in 2024, as we commemorate the 20th anniversary of graphene's experimental discovery, it is an opportune moment to review on past achievements and to anticipate future in this field.

Graphene is renowned for its extraordinary properties and vast potential across a wide range of applications. First, it has a unique band structure, where the conduction and valence bands meet at the Dirac points, giving rise to massless Dirac fermions with velocities approximately 1/300 of the speed of light. This linear dispersion relation near the Dirac points leads to an unusual filling factor sequence for the quantum Hall effect (QHE) and large

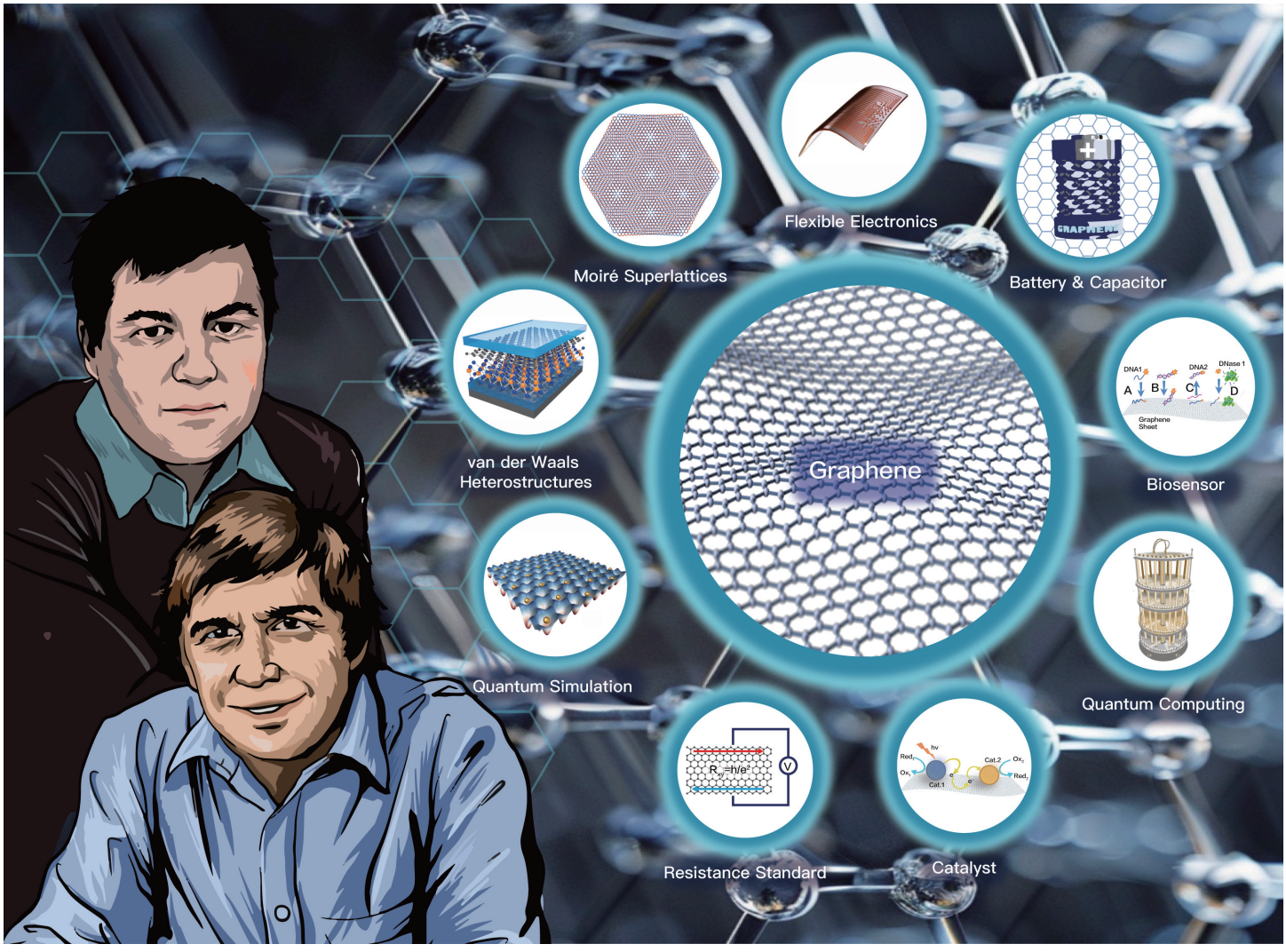

Figure 1. The 2010 Nobel Laureates in Physics (Konstantin Novoselov and Andre Geim), and the emerging applications of graphene.

quantum Hall gaps for the low Landau levels, allowing for the observation of the QHE at room temperature. International metrology organizations have also recognized the potential of the QHE in graphene for defining resistance standards.[2] Meanwhile, graphene exhibits a high level of electrical tunability, thermal conductivity, transparency, and flexibility, combined with a high mechanical strength and light weight, making it an ideal material for many applications, ranging from flexible electronics and high-frequency transistors to transparent conductive coatings and advanced composites. In energy storage, graphene-enhanced batteries and supercapacitors offer the potential for faster charging and greater capacity. Graphene's biocompatibility has

also opened research avenues in biotechnology and medicine, including drug delivery systems, biosensors, and tissue engineering. Its high surface area and conductive properties are being leveraged in environmental applications, such as water purification and sensors for detecting pollutants and other hazardous substances.

In the early days of graphene research, silicon wafers with SiO_2 coating are typically used as the substrates. However, the roughness and charged impurities of SiO_2 surfaces led to significant scattering of charge carriers, thereby limiting the electron mobility of graphene to about ~10, 000 cm^2/Vs. To attain atomically flat surfaces, methods like suspension or the use of inherently smoother substrates, such as mica and hexagonal boron nitride (hBN), have been introduced. Especially, the encapsulation with hBN, a layered insulating material notable for its large band gaps and absence of dangling bonds, has been shown to significantly enhance graphene's mobility and enable high electronic performance. At the room temperature, the mobility can achieve purely phonon limited (well above ~20, 000 cm^2/Vs) and outperforms all the other materials. While at lower temperatures, many intrinsic electronic properties like ballistic transport (mean free path >10 μm), electronic hydrodynamics, and fractional QHE become easily accessible. In the pursuit of enhancing graphene's mobility, the development of the dry-transfer method has shown broad implications for the field.[3] This technique, harnessing the van der Waals (vdW) forces between layers without necessitating direct contact with organics or solvents, has now been widely adopted in the fabrication of various vdW heterostructures, facilitating the exploration of new physics and device functionalities for an array of 2D materials, including semiconductors, superconductors, and magnets.

Twisted bilayer graphene (TBG) has emerged as another significant milestone since the discover of graphene. It is known that correlation effects are generally weak for monolayer graphene with linearly dispersing band structures, owing to the large bandwidths relative to Coulomb interactions. Both theoretical and experimental studies have shown that overlaying two graphene layers with a small rotation angle ~1.1° — known as the magic angle — can remarkably alter the electronic structure and result in a flat-band condition. Such band flattening is pivotal for enhancing electron correlations. Since 2018, many intriguing emergent quantum states including the unconventional superconductivity, correlated insulators, orbital magnetism, and quantum anomalous Hall effect have been discovered in the TBG. The idea of overlaying two materials with a slight lattice or angular mismatch to form moiré superlattices has now greatly expanded beyond TBG and opened an entirely new research avenue. These moiré superlattices, characterized by a long-wavelength (typically around 10nm) and periodically modulating real-space lattice stacking registry, offer a versatile platform for quantum simulations that probe many-body and topological physics.

Besides TBG and other graphene-based moiré superlattices, the flat bands can also be realized in rhombohedral graphene, such as in its multilayer configurations like the trilayer, tetralayer, and pentalayer. This form of graphene is distinguished by its stacking order being different from the usual Bernal (AB) stacked structures, resulting in a topological electronic structure where energy bands become progressively flatter with an increase in the number of layers. Very recently, emergence of exotic phenomena like isospin-polarized metals, orbital multiferroicity, integer and even fractional quantum anomalous Hall effects have been reported for multilayered rhombohedral graphene or its moiré structures.[4] These states can potentially be used for developing ultralow-power electronics for information storage, transportation, and fault-tolerant topological quantum computation.

Despite numerous advances in the graphene research, both its fundamental study and broad market application encounter several key challenges. Nevertheless, some notable progress has been made. First, due to the small size of high-quality devices, many experimental methods that are developed for studying bulk materials cannot be easily applied to graphene. However, recently, we've witnessed several innovations in the methodology of studying graphene devices and its related structures, for example, the STM (scanning tunneling microscopy) for visualizing various symmetry breaking states, micro/nano- ARPES (angle-resolved photoemission spectroscopy) for resolving the band structures, the scanning SQUID (superconducting quantum interference device)-on-tip (SOT) microscopy for measuring small local magnetic signals, and the Rydberg exciton sensing technique for optically detecting the electronic compressibility.[5] Second, many emergent phenomena in graphene and its moiré superlattices remain poorly understood, such as the pairing mechanism for the superconductivity, the composite Fermi liquids, and the possible non-abelian states. Third, TBG has encountered reproducibility challenges owing to the difficulties in fabricating large-area samples with precise rotational alignment and free of spatial inhomogeneities. On the application side, scalable production, cost of high-quality graphene and the integration into existing manufacturing processes and materials poses great challenges. In recent years, methods like chemical vapor deposition (CVD) have been demonstrated to be capable in producing meter-sized single-crystal graphene, showing opportunities in solving some of the above problems. On the other hand, how to introduce substantial band gaps into graphene for semiconductor applications remains a long-standing challenge. Various strategies have been extensively explored, including chemical doping, production of graphene nanoribbons, and substrate engineering for gap opening in bilayer graphene. Recently, epitaxial graphene on silicon carbide has provided a hopeful pathway for realizing high-mobility semiconducting graphene.

In summary, two decades after graphene's isolation, its evolution from a theoretical concept to a cornerstone in material science has been marked by both significant breakthroughs and formidable challenges. This era has witnessed the introduction of innovative techniques for the production, device fabrication, and measurements of graphene, alongside groundbreaking discoveries like magic-angle TBG and moiré superlattices. These advancements have not only deepened our understanding of quantum phenomena but also expanded the scope of potential applications. The field of graphene is now poised on the brink of further transformative developments in future science and technology.

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DECLARATION OF INTERESTS

The authors declare no competing interests.