

Recent Progress on Two-Dimensional Materials

Cheng Chang¹, Wei Chen², Ye Chen³, Yonghua Chen⁴, Yu Chen⁵, Feng Ding⁶, Chunhai Fan⁷, Hong Jin Fan⁸, Zhanxi Fan⁹, Cheng Gong¹⁰, Yongji Gong¹¹, Qiyuan He¹², Xun Hong¹³, Sheng Hu¹⁴, Weida Hu¹⁵, Wei Huang⁴, Yuan Huang¹⁶, Wei Ji¹⁷, Dehui Li¹⁸, Lain-Jong Li¹⁹, Qiang Li²⁰, Li Lin²¹, Chongyi Ling²⁰, Minghua Liu²², Nan Liu²³, Zhuang Liu²⁴, Kian Ping Loh², Jianmin Ma²⁵, Feng Miao²⁶, Hailin Peng²⁷, Mingfei Shao²⁸, Li Song²⁹, Shao Su³⁰, Shuo Sun³¹, Chaoliang Tan³², Zhiyong Tang³³, Dingsheng Wang³⁴, Huan Wang³⁵, Jinlan Wang²⁰, Xin Wang³⁶, Xinran Wang³⁷, Andrew T. S. Wee³¹, Zhongming Wei³⁸, Yuen Wu³⁶, Zhong-Shuai Wu³⁹, Jie Xiong⁴⁰, Qihua Xiong⁴¹, Weigao Xu⁴², Peng Yin⁴³, Haibo Zeng⁴⁴, Zhiyuan Zeng¹², Tianyou Zhai⁴⁵, Han Zhang⁴³, Hui Zhang⁴, Qichun Zhang¹², Tierui Zhang⁴⁶, Xiang Zhang⁴⁷, Li-Dong Zhao¹¹, Meiting Zhao⁴⁸, Weijie Zhao²⁰, Yunxuan Zhao⁴⁶, Kai-Ge Zhou⁴⁹, Xing Zhou⁴⁵, Yu Zhou⁵⁰, Hongwei Zhu⁵¹, Hua Zhang^{9,*}, Zhongfan Liu^{27,*}

¹ Institute of Science and Technology Austria, Am Campus 1, 3400 Klosterneuburg, Austria.

² Department of Chemistry, National University of Singapore, 3 Science Drive 3, 117543, Singapore.

³ Department of Chemistry, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong, China.

⁴ Key Laboratory of Flexible Electronics and Institute of Advanced Materials, Nanjing Tech University, Nanjing 211816, China.

⁵ School of Life Sciences, Shanghai University, Shanghai 200444, China.

⁶ Centre for Multidimensional Carbon Materials, Institute for Basic Science, Ulsan 44919, Korea.

⁷ School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China.

⁸ School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore.

⁹ Department of Chemistry, City University of Hong Kong, Kowloon, Hong Kong, China.

¹⁰ Department of Electrical and Computer Engineering and Quantum Technology Center, University of Maryland, College Park, Maryland 20742, USA.

¹¹ School of Materials Science and Engineering, Beihang University, Beijing 100191, China.

¹² Department of Materials Science and Engineering, City University of Hong Kong, Kowloon, Hong Kong, China.

¹³ Center of Advanced Nanocatalysis (CAN), Hefei National Laboratory for Physical Sciences at the Microscale, Department of Applied Chemistry, University of Science and Technology of China, Hefei 230026, China.

¹⁴ College of Chemistry and Chemical Engineering, State Key Laboratory of Physical Chemistry of Solid Surfaces, Collaborative Innovation Center of Chemistry for Energy Materials (iChEM), Xiamen University, Xiamen, 361005, Fujian Province, China.

¹⁵ State Key Laboratory of Infrared Physics, Shanghai Institute of Technical Physics, Chinese Academy of Sciences, Shanghai 200083, China.

¹⁶ Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing, 100081, China.

¹⁷ Beijing Key Laboratory of Optoelectronic Functional Materials & Micro-Nano Devices, Department of Physics, Renmin University of China, Beijing 100872, China.

¹⁸ School of Optical and Electronic Information, Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan 430074, China.

¹⁹ Department of Mechanical Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong, China.

²⁰ School of Physics, Southeast University, Nanjing 211189, China.

²¹ Department of Materials Science and Engineering, National University of Singapore, Singapore 117575, Singapore.

²² CAS Key Laboratory of Colloid, Interface and Chemical Thermodynamics, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China.

²³ College of Chemistry, Beijing Normal University, Beijing 100875, China.

²⁴ Institute of Functional Nano & Soft Materials (FUNSOM), Jiangsu Key Laboratory for Carbon-based Functional Materials and Devices, Soochow University, Suzhou 215123, Jiangsu Province, China.

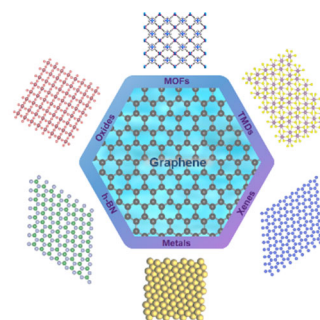
²⁵ School of Materials and Energy, University of Electronic Science and Technology of China, Chengdu 611731, China.

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*Corresponding authors. Emails: Hua.Zhang@cityu.edu.hk (H.Z.); zfliu@pku.edu.cn (Z.L.).

- ²⁶ School of Physics, Nanjing University, Nanjing 210093, China.
- ²⁷ Center for Nanochemistry (CNC), Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Beijing Graphene Institute (BGI), Peking University, Beijing 100871, China.
- ²⁸ State Key Laboratory of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 100029, China.
- ²⁹ National Synchrotron Radiation Laboratory, CAS Center for Excellence in Nanoscience, University of Science and Technology of China, Hefei 230029, China.
- ³⁰ State Key Laboratory of Organic Electronics and Information Displays & Jiangsu Key Laboratory for Biosensors, Institute of Advanced Materials (IAM), Nanjing University of Posts and Telecommunications, Nanjing 210023, China.
- ³¹ Department of Physics, National University of Singapore, 2 Science Drive 3, 117551, Singapore.
- ³² Department of Electrical Engineering, City University of Hong Kong, Kowloon, Hong Kong, China.
- ³³ CAS Key Laboratory of Nanosystem and Hierarchical Fabrication, CAS Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China.
- ³⁴ Department of Chemistry, Tsinghua University, Beijing 100084, China.
- ³⁵ Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), Renewable Energy Conversion and Storage Center (RECAST), College of Chemistry, Nankai University, Tianjin 300071, China.
- ³⁶ School of Chemistry and Materials Science, iChEM (Collaborative Innovation Center of Chemistry for Energy Materials), Hefei National Laboratory for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei 230026, China.
- ³⁷ National Laboratory of Solid State Microstructures, School of Electronic Science and Engineering, Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210023, China.
- ³⁸ Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China.
- ³⁹ State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, Liaoning Province, China.
- ⁴⁰ State Key Laboratory of Electronic Thin Film and Integrated Devices, University of Electronic Science and Technology of China, Chengdu 610054, China.
- ⁴¹ State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, Tsinghua University, Beijing 100084, China.
- ⁴² Key Laboratory of Mesoscopic Chemistry, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing 210023, China.
- ⁴³ Institute of Microscale Optoelectronics, Shenzhen University, Shenzhen 518060, Guangdong Province, China.
- ⁴⁴ MIIT Key Laboratory of Advanced Display Materials and Devices, College of Material Science and Engineering, Nanjing University of Science and Technology, Nanjing 210094, China.
- ⁴⁵ School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, China.
- ⁴⁶ Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China.
- ⁴⁷ Faculties of Sciences and Engineering, The University of Hong Kong, Hong Kong, China.
- ⁴⁸ Tianjin Key Laboratory of Molecular Optoelectronic Sciences, Department of Chemistry, Institute of Molecular Aggregation Science, Tianjin University, Tianjin 300072, China.
- ⁴⁹ Institute of Molecular Plus, Tianjin University, Tianjin 300072, China.
- ⁵⁰ School of Physics and Electronics, Hunan Key Laboratory of Nanophotonics and Devices, Central South University, Changsha 410083, China.
- ⁵¹ State Key Lab of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China.

Abstract: Research on two-dimensional (2D) materials has been explosively increasing in last seventeen years in varying subjects including condensed matter physics, electronic engineering, materials science, and chemistry since the mechanical exfoliation of graphene in 2004. Starting from graphene, 2D materials now have become a big family with numerous members and diverse categories. The unique structural features and physicochemical properties of 2D materials make them one class of the most appealing candidates for a wide range of potential applications. In particular, we have seen some major breakthroughs made in the field of 2D materials in last five years not only in developing novel synthetic methods and exploring new structures/properties but also in identifying innovative applications and pushing forward commercialisation. In this review, we provide a critical summary on the recent progress made in the field of 2D materials with a particular focus on last five years. After a brief background



introduction, we first discuss the major synthetic methods for 2D materials, including the mechanical exfoliation, liquid exfoliation, vapor phase deposition, and wet-chemical synthesis as well as phase engineering of 2D materials belonging to the field of phase engineering of nanomaterials (PEN). We then introduce the superconducting/optical/magnetic properties and chirality of 2D materials along with newly emerging magic angle 2D superlattices. Following that, the promising applications of 2D materials in electronics, optoelectronics, catalysis, energy storage, solar cells, biomedicine, sensors, environments, *etc.* are described sequentially. Thereafter, we present the theoretic calculations and simulations of 2D materials. Finally, after concluding the current progress, we provide some personal discussions on the existing challenges and future outlooks in this rapidly developing field.

Key Words: Two-dimensional materials; Transition metal dichalcogenides; Phase engineering of nanomaterials; Electronics; Optoelectronics; Catalysis; Energy storage and conversion

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1 Introduction

The first report on the mechanical cleavage of atomically thin single-crystalline carbon film, namely graphene, and its extraordinary transport properties by Geim, Novoselov and co-workers¹ in 2004 had ignited the resurgence of a class of fascinating functional nanomaterials, *i.e.*, two-dimensional (2D) materials²⁻¹⁰. 2D materials now have been recognized as a type of nanomaterials which have a sheet-like morphology featuring with a large lateral size from hundreds of nanometers to tens of micrometers or even larger but a thickness in single or few atomic layer^{2,3}. Such a unique structural feature of 2D materials

endows them with various unconventional physical, chemical, optical, electronic and magnetic properties as compared to their bulk, zero-dimensional (0D) and one-dimensional (1D) counterparts². Owing to their unusual properties, 2D materials have been proven to be one of the most promising candidates for numerous potential applications like electronics^{11–14}, optoelectronics^{15–20}, catalysis^{21–26}, energy storage^{27–34}, solar cells^{35–38}, biomedicine^{39–45}, sensors^{46–49}, environments^{50–54}, *etc.* Driven by their unusual properties and promising applications, a large number of novel 2D materials beyond graphene, such as transition metal dichalcogenides (TMDs) including MoS₂, MoSe₂, MoTe₂, WS₂, WSe₂, ReS₂, TaS₂, *etc.*)^{55,56}, hexanol boron nitride (h-BN)^{57,58}, graphyne^{59,60}, noble metal dichalcogenides (NMDs: PdSe₂, PtSe₂, PtS₂, *etc.*)^{61,62}, elemental 2D materials (e.g., black phosphorus (BP), tellurium, silicene, germanene, borophene, *etc.*)^{63–65}, layered metal oxides^{66,67}, layered double hydroxides (LDHs)^{68,69}, graphitic carbon nitride (g-C₃N₄)^{70,71}, MXenes⁷², metals⁷³, organics/polymers^{74,75}, metal-organic frameworks (MOFs)^{76,77}, covalent-organic frameworks (COFs)^{78,79}, organic-inorganic hybrid perovskites^{80–82}, and transition metal halides^{83,84}, have been synthesized by various synthetic methods in the last decade. It is worth pointing out that the number of materials in the family of 2D materials is still continuously growing every year.

On the basis of the previous research works, the last five years have witnessed some major breakthroughs made in the field of 2D materials in all aspects. Firstly, a large number of novel 2D materials have been reported, including NMDs^{85–87}, tellurium^{88,89}, selenium⁹⁰, and so on. Secondly, some novel methods have been developed for synthesis of 2D materials with higher quality, larger size, or better control, such as oxygen plasma- or Au-enhanced mechanical exfoliation^{91,92}, organic intercalation-assisted liquid exfoliation of layered materials (e.g., BP, TMDs, InSe, *etc.*)^{93–95}, salt-assisted chemical vapor deposition (CVD) growth of a library of 2D thin layers^{96,97}, CVD growth of wafer-scale high-quality 2D thin films^{98,99}, pulsed laser deposition (PLD) of BP thin films¹⁰⁰, vapor phase synthesis of high-purity 1T'-phase 2D TMD crystals¹⁰¹, and liquid metal-assisted synthesis of metal oxide nanosheets¹⁰². Thirdly, some new promising applications of 2D materials have been demonstrated, such as integrated circuits based on wafer-scale 2D thin films¹⁰³ and infrared imaging sensor systems based on graphene¹⁰⁴. More importantly, some newly emerging research directions have been extensively explored on 2D materials in recent years. For example, the phase engineering of nanomaterials (PEN) including 2D materials has been recognized as a promising way to fine tune their physicochemical properties and enhance their performances in addition to other conventional structural characteristics, such as size, thickness, defects, vacancies, and interlayer spacing^{105,106}. As another typical example, by simply stacking two 2D graphene in a specific magic angle, namely magic angle 2D superlattices, the properties of graphene can be tuned from a conductor to a superconductor or insulator^{107,108}.

Inspired by the unexpected properties of magic angle graphene superlattices, magic angle 2D superlattices have become one of the most interesting materials to explore new properties in condensed matter physics.

Although many review articles related to 2D materials have been published previously, most of them were published in several years ago or even earlier, and most of them focused on a selected type 2D materials (e.g., graphene, graphyne, TMDs, MOFs, elemental ones, metals, MXenes, *etc.*)^{4,55,73}, or specific application (e.g., electronics, optoelectronics, energy storage, electrocatalysis, sensors, biomedicine, *etc.*)^{27,31,39}. Bearing this mind, offering a comprehensive review article to cover all of the 2D materials from all aspects with highlights on recent progress in this growing field is of great significance for its further development. To this end, this review aims to critically summarize the recent progress on 2D materials with particular focus on the last five years. Following a brief background introduction, the major synthetic methods for 2D materials, including the mechanical exfoliation, liquid exfoliation, vapour phase deposition, and wet-chemical synthesis as well as phase engineering of 2D materials are first discussed. The superconducting, optical, magnetic properties and chirality of 2D materials along with newly emerging magic angle 2D superlattices are then introduced. Thereafter, we summarize the great potential of 2D materials in various applications like electronics, optoelectronics, catalysis, energy storage, solar cells, biomedicine, sensors, environments, *etc.* Following that, recent progress on the theoretic calculations and simulations of 2D materials is also discussed. Finally, we conclude this Review by summarizing the current process and offering some personal insights on the existing challenges and future opportunities in this promising field.

2 Synthetic methods

2.1 Mechanical exfoliation

Mechanical exfoliation has been recognized as an efficient method to obtain fresh atomically flat surface of layered materials^{109,110}. In 2004, a new tape-based exfoliation method developed in Geim's group was used to prepare monolayer and few-layer graphene from graphite^{1,111}. As one of the most popular "top-down" strategy to date, this mechanical exfoliation technique has been widely used to get a large number of 2D crystals, such as MoS₂^{112,113}, WS₂¹¹⁴, SnS₂¹¹⁵ and BP^{115,116}. The exfoliated 2D materials are ideal samples to study their intrinsic electronic^{117,118}, optical¹¹⁹ and mechanical properties. However, there are a few shortcomings for the conventional mechanical exfoliation. Firstly, the sample size of exfoliated 2D materials is usually in the range of few to tens of micrometers. Secondly, the yield is quite low. With the discovery of new layered materials, novel mechanical methods are desired to prepare high-quality, large-area 2D materials with relatively high efficiency.

Although the exfoliation processes are generally simple, some scientific questions were not well understood at the beginning.