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Overview

The successful exfoliation of monolayer graphene has triggered the eruptive research and development efforts of two-dimensional (2D) materials in the format of monolayer, bilayers, few layers, and heterostructure. With properties distinct from their bulk counterparts, 2D materials enable tightly confined light and phonons, unprecedentedly controlled electrons, spins and excitons, which evoke fundamental new science and offer new paradigm technologies for highly integrated multifunctional optoelectronic devices. In this editorial, we briefly review the interesting new advances in the past few years and highlight the remaining challenges and identified opportunities. As a dedicated research journal serving the photonics community, APL Photonics eagerly looks forward to seeing more exciting findings in the 2D material photonics area to be disseminated in such an excellent platform over the coming years.

I. INTRODUCTION

The discovery of graphene\(^1\) has spawned enthusiastic research into other atomically thin materials.\(^1\) To date, many different materials have been successfully exfoliated which significantly expand the available options to achieve the desired optoelectronic properties and extend the wavelengths of interest. A suite of atomically thin allotropes and compounds has been intensively studied, including semimetals (graphene), insulators (e.g., hexagonal boron nitride), semiconductors (e.g., transition metal dichalcogenides, TMDs), and additional semimetals (e.g., black phosphorus). Research on atomically thin layers of metals such as palladium and rhodium has also been reported.\(^1\) In addition, there is growing interest in layered compounds, for example, inorganic perovskites.\(^2\) These 2D materials have offered extraordinary electronic, chemical, optical, magnetic, mechanical, and thermal properties,\(^2\) which give birth to many new burgeoning research fields based on 2D materials, including material simulation and design, synthesis and manufacturing, characterization, and device design and fabrication.

As far as photonics is concerned, the natural strong light-material interactions enabled by 2D materials are highly preferred for achieving desired functionalities in miniaturized photonics systems. For example, nanometer thickness ultrathin flat lenses even down to a monolayer level\(^5\)–\(^7\) have been demonstrated in graphene, graphene oxide (GO), and TMDs due to strong light modulations. High nonlinearity that is several orders of magnitude larger than the conventional semiconductors has been achieved, which enables high performance saturable absorbers for ultrafast modelocking and ultracompact nonlinear photonic chips.\(^8\)–\(^10\) More importantly, by tuning the carrier density or optical bandgap via electrical or optical means, the optical properties of 2D materials can be instantly changed, making them versatile for diverse in situ tunable optical devices. The advances in material species exploration allow the realization of multiple functions by stacking layers of different 2D materials into heterostructures to form ultimately integrated optoelectronic devices holding great promise to succeed the legacy of silicon.

Although it has been almost 15 years since it was discovered,\(^1\) graphene and its family of materials, such as graphene oxide, are still garnering significant research interest. It is expected that it will remain a research focus for years to come. Advances in the understanding of fundamental properties and synthesis and fabrication methods have paved the way for the development of various graphene-based applications and functional devices, including light sensing, harvesting, emitting devices, transparent conductors, optical modulators, spintronics, energy storage devices, biomedical applications, and communication devices. In particular, the recent advances in 2D materials’ nonlinearity has ignited many new explorations of fundamental understandings and provided new ideas on device integration. APL Photonics featured a special topic
on “Nonlinear Optics in 2D Materials” in March 2019 to capture advances in this flourishing field.

Graphene is considered as a record-performance nonlinear-optical material on the basis of numerous experiments. The optical nonlinearity of graphene family materials has been reported extensively. From the theoretical aspect, recently, a distant-neighbor quantum-mechanical (DNQM) approach was proposed to study the linear and nonlinear optical properties of graphene nanoflakes (GNFs). By embedding a cavity into GNFs, one can change their symmetry properties, tune their optical properties, or enable otherwise forbidden second-harmonic generation processes. In the meantime, third-harmonic generation (THG) can be widely tuned in graphene using an electric gate voltage. The demonstration of broadband, electrically tunable third-order nonlinear optical responses in graphene is promising. In a recent theoretical work, a complex phenomenon called saturable photoexcited-carrier refraction was proposed to be at the heart of nonlinear-optical interactions in graphene such as self-phase modulation. This work sheds light on the understanding of 2D-material nonlinearities and finally enables their full exploitation in next-generation nonlinear-optical devices. Exciting experimental progress of enhanced four-wave mixing (FWM) in doped silica waveguides integrated with well-defined graphene oxide (GO) layers was reported recently. Strong mode overlap between the integrated waveguides and the GO films enables a high Kerr nonlinearity and low loss, which significantly improved the FWM efficiency of the hybrid integrated waveguides.

In the terahertz region, graphene is a promising magneto-optical material. However, high Faraday rotation is only achievable at a low terahertz frequency range in single layer graphene. Simultaneous Faraday rotation and optical transmission enhancement in Au grating/graphene/silicon hybrid plasmonic structures across a wide frequency range using extraordinary transmission of terahertz spoof surface plasmons was demonstrated. In a broad frequency range up to 13.1 THz, the Faraday rotation and magneto-optical figure of merit in this hybrid structure can exceed the maximum value of single layer graphene at the low terahertz frequency range, providing a possible candidate for magneto-optical systems.

Investigating material species other than graphene expands the options available for meeting the desired optoelectronic properties and wavelengths of interest. One example of a new 2D optical material is the transition metal dichalcogenides (TMDCs). Layered 2D materials with extraordinary optical properties play important roles in the development of ultrafast photonics, in which mode-locking lasers with a high fundamental repetition rate (>1 GHz) are of particular interest. The nonlinear optical properties of one of the emerging 2D materials, rhenium diselenide (ReSe$_2$), have been found to have broadband ultrafast saturable absorption of ReSe$_2$ from visible to the near infrared wavelength regimes, which enables potential applications in ultrafast lasing.

Single layer molybdenum diselenide from the family of TMDCs can have unexpected high levels of reflectance, which can be made into mirrors that can reflect a considerable proportion of the incident light—up to 85% at the exciton resonance frequency of the material. This finding could have technological implications for nanophotonics, optoelectronics, and quantum optics. TMDC monolayers have naturally terminated surfaces and can exhibit a near-unity photoluminescence quantum yield in the presence of suitable defect passivation. To date, steady-state monolayer light-emitting devices suffer from Schottky contacts or require complex heterostructures. A transient-mode electroluminescent device based on transition-metal dichalcogenide monolayers (MoS$_2$, WSe$_2$, MoSe$_2$, and WSe2) can overcome these problems. Electroluminescence from the dopant-free two-terminal device was obtained by applying an AC voltage between the gate and the semiconductor. The electroluminescence intensity was shown to be weakly dependent on the Schottky barrier height or polarity of the contact.

Infrared photodetectors are seeing rapid growth in the application space, with an increasing demand for compactness, sensitivity, and cost-effectiveness. The main limiting factors from more widespread applications of such devices lie in the need for cooling and the high costs associated with the processing of high-quality semiconductors. Black phosphorous (BP)/MoS$_2$ heterojunction photodiodes have been explored as mid-wave infrared (MWIR) detectors with significantly improved external quantum efficiencies reaching 35% at the room temperature. By leveraging the anisotropic optical properties of BP, bias-selectable polarization-resolved photodetection was demonstrated. On the other hand, bilayers of PtSe$_2$ combined with defect modulation possess strong light absorption in the mid-infrared region, leading to a photoconductive detector operating in a broadband due to a variable bandgap. An infrared photodetector based on 2D Bi$_2$O$_2$Se crystal has demonstrated both high sensitivity and ultrafast response at the room temperature. Recently, the demonstration of patterned graphene metamaterials may provide new solutions for IR detectors.

II. CHALLENGES AND OPPORTUNITIES

To further advance the 2D materials in the optical applications, there are several challenges that need to be addressed in the near future.

A. Developing computational methods to predict optical properties of 2D materials

The ability of 2D materials to absorb, reflect, and act upon light is of primary importance to develop optoelectronics and optical devices that interact with light to achieve functionalities. Computational methods are critical to understand how the atom-thin materials refine the modulation and manipulate the light. The development of such a theoretical framework is important to evaluate any 2D material at a deeper quantitative level according to the atomic structure, and guide the design and manipulation of 2D materials for desired optical properties. In addition, it is expected that theoretical methods, which are able to model 2D material stacks—van der Waals heterostructures—could lead to significant impact in 2D material research through broadening the spectral range and bringing new functionalities.

B. Large scale synthesis of 2D materials and van der Waals heterostructures

Currently, most 2D materials are synthesized by the relatively simple chemical vapor deposition (CVD) method, which do not always provide precise control over the grown materials and can often result in limited homogeneity. The resulting films usually display properties inferior to those of mechanically exfoliated...
layers. Therefore, there is an urgent need in synthesizing high-quality, large-area 2D materials in various species towards industrial fabrication (wafer scale) and device assembly. There are demonstrations of low cost solution based synthetic methods, which occur at much lower temperatures and without high vacuum. However, the controllability of the process and the quality of the resulting 2D materials are generally lower than those obtained through CVD and mechanical exfoliation methods. Further improvements in solution based synthetic methods with better control and quality may lead to important breakthroughs in the applicability of 2D materials.

2D heterostructures are mostly developed by directly stacking individual monolayer flakes of different materials. Although this method allows ultimate flexibility, it is slow and cumbersome. Thus, techniques involving transfer of large-area crystals grown by CVD, direct growth of heterostructures by CVD or physical epitaxy, or one-step growth in solution see a great demand. The rapid progress of graphene technology over the past few years signals positively the upcoming boom of 2D heterostructures with potential in both fundamental breakthroughs and technological advancements.

C. Improve the stability of 2D materials in ambient and working conditions

While 2D materials host many exciting potential applications, some of these 2D materials are subject to environmental instability induced by interactions with gas molecules in air, which poses a barrier to practical applications and scalable manufacture. To overcome this issue, it is necessary to understand the origin of material instability and interaction with molecules commonly found in air as well as developing a reproducible and manufacturing compatible method to postprocess these materials to extend their lifetime. There have been a number of demonstrations, which can significantly enhance their environmental stability through physical protection, chemical functionalization, or passivation. As a result, more air stable 2D materials are expected in the coming years.

In the meantime, it is more important to improve the stability of 2D materials in working conditions. Due to the ultrathin nature, 2D materials can be easily degraded or damaged by high fluence of photons. Take optical nonlinearity application for example, the optical nonlinear effect is always accompanied by an inevitable linear photodissociation, or passivation. As a result, more air stable 2D materials are expected in the coming years.

In addition, it will be exciting to see more work on the optical applications of van der Waals heterostructures. Another attractive frontier is 2D material based plasmonics. Compared with surface plasmons in typical metallic structures, 2D materials can offer much tighter optical confinement due to its atomic layer thickness. This significantly enhanced light–matter interaction makes it possible to greatly reduce the amount of material and optical power required for light manipulation, and thus minimizing the footprint and power consumption of optical devices. Other opportunities in this field may come from those 2D materials that have not yet been used for optical applications.

D. Achieving tunability of 2D materials

The optical properties of 2D materials possess unique tunability through controlling the doping density and type of ions, defects levels, local excitonic effect, structural response, and the charge transfer in 2D materials. Recently, electrolyte gating has been used as a tool to unlock exciting new physics in 2D materials. Moreover, ions can be expected to intercalate between the layers of the 2D crystals—a mechanism that is well studied by the battery community. For optical applications, dynamic electrical or optical tuning is preferred. However, the optical properties (i.e., absorption) and spatial distribution (the spatial distribution may affect the phase and amplitude distributions of lights going through the system) of electrolytes may significantly affect the optical performance of the entire system. In addition, it is challenging to characterize the optical constants (refractive index and extinction coefficient) of the whole system when the sample sizes are limited to microscopic. It will be exciting to see more work on light-driven tunable optical devices with 2D materials.

III. CONCLUSIONS AND OUTLOOK

In conclusion, the time from the discovery of a new material to large-scale commercialization usually takes 20–30 years. Compared with traditional materials, 2D materials are novel and almost every day we see interesting publications on them across a variety of disciplines. Accompanied with the advancement in developing computational methods to predict optical properties of 2D materials, large-scale synthesis of 2D materials and van der Waals heterostructures, improving the stability of 2D materials in ambient and working conditions and tuning the properties of 2D materials, their development roadmap, and commercialization pathways can possibly be identified with many applications for different 2D materials in various industries. Collectively, the family of 2D materials is sure to play an important role in our society. As a leading photonics journal, APL Photonics is eager to consider papers addressing the abovementioned challenges.

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