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To cite this article: K Saumya *et al* 2023 *Nano Futures* **7** 032005

NANO FUTURES

TOPICAL REVIEW

'Magic' of twisted multi-layered graphene and 2D nano-heterostructures

K Saumya[1], S Naskar[2] and T Mukhopadhyay[2,*]

[1] Department of Aerospace Engineering, Indian Institute of Technology Kanpur, Kanpur, India
[2] Faculty of Engineering and Physical Sciences, University of Southampton, Southampton, United Kingdom
* Author to whom any correspondence should be addressed.

E-mail: t.mukhopadhyay@soton.ac.uk

Keywords: twisted bilayer graphene, magic angle, van der Waals heterostructures, superconductivity, 2D materials, twistronics

Supplementary material for this article is available online

OPEN ACCESS

RECEIVED
1 April 2023

REVISED
3 July 2023

ACCEPTED FOR PUBLICATION
15 August 2023

PUBLISHED
5 September 2023

Abstract

Two-dimensional materials with a single or few layers are exciting nano-scale materials that exhibit unprecedented multi-functional properties including optical, electronic, thermal, chemical and mechanical characteristics. A single layer of different 2D materials or a few layers of the same material may not always have the desired application-specific properties to an optimal level. In this context, a new trend has started gaining prominence lately to develop engineered nano-heterostructures by algorithmically stacking multiple layers of single or different 2D materials, wherein each layer could further have individual twisting angles. The enormous possibilities of forming heterostructures through combining a large number of 2D materials with different numbers, stacking sequences and twisting angles have expanded the scope of nano-scale design well beyond considering only a 2D material mono-layer with a specific set of given properties. Magic angle twisted bilayer graphene (BLG), a functional variant of van der Waals heterostructures, has created a buzz recently since it achieves unconventional superconductivity and Mott insulation at around 1.1° twist angle. These findings have ignited the interest of researchers to explore a whole new family of 2D heterostructures by introducing twists between layers to tune and enhance various multi-physical properties individually as well as their weighted compound goals. Here we aim to abridge outcomes of the relevant literature concerning twist-dependent physical properties of BLG and other multi-layered heterostructures, and subsequently highlight their broad-spectrum potential in critical engineering applications. The evolving trends and challenges have been critically analysed along with insightful perspectives on the potential direction of future research.

1. Introduction

One of the most fascinating materials with superior nano-scale properties, graphene has always been an interest to researchers all over the globe since its discovery by micro-mechanical exfoliation of bulk graphite crystals using a scotch tape [1–5]. Its one atomic thick two dimensional (2D) nature gives rise to some exceptional phenomena such as ultra-high electron mobility [6, 7], superior thermal conductivity [8], unconventional quantum Hall effect [9], generation of massless Dirac fermions [10, 11] etc along with extraordinary mechanical properties in terms of strength and Young's modulus [12–14]. Graphene and its different derivatives along with the effect of defects and doping have been at the centre of attraction over the last decade for exploring various multifunctional properties [15–24]. The physical properties vary significantly when two or more layers come into play (referred to as nano-heterostructure [25–27]) due to interlayer coupling, stacking sequences, and twisting. The current paper focuses on achieving exciting and unprecedented multi-physical properties through stacking and twisting graphene layers along with the evolving trends of expanding the functional design scope through introducing other 2D materials [28–30] to form twisted nano-heterostructures (refer to figure 1).

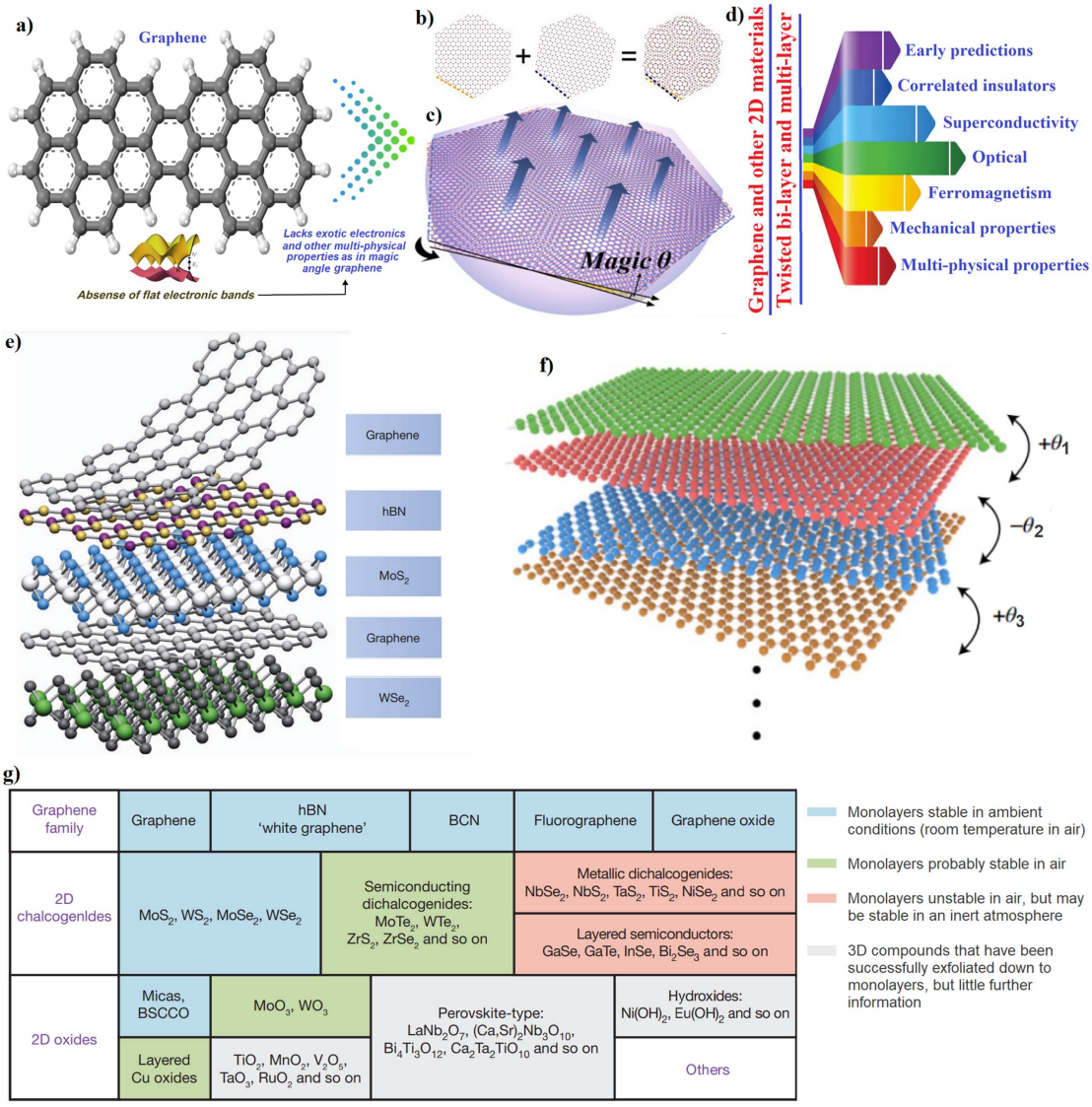

Figure 1. Introductory overview of the nano-architecture of Moiré superlattices consisting of 2D materials. (a) Single layer graphene (lacking flat electronic bands). (b), (c) Twisted bi-layers [31], where the twisted architecture in a bi-layer graphene is schematically shown. Reprinted from [31], © 2020 Elsevier Inc. (d) Multi-physical properties that can be modulated in twisted 2D materials. (e),(f) Multi-layered architectures of graphene and other 2D materials. Note that it is possible to have multi-layered graphene with different twist angles at each layers [32]. Reproduced from [32], Copyright © 2022, The Author(s), under exclusive licence to Springer Nature Limited. Further, graphene and other 2D material heterostructures (including mono-planar and multi-planar nanostructural configuration [33] of 2D materials in each layer) can have different twist angles at each layers, leading to a wide range of physical properties [34]. (g) Different 2D materials [34] that can potentially constitute the layers of a heterostructure with or without layer-wise twist. The focus of this paper is to systematically review different physical properties (as shown in subfigure (d)) for twisted bi-layer graphene and different other twisted heterostructures in an expanded design space of number of layers, different 2D materials and their respective twisting angle. Reproduced from [34], Copyright © 2013, Springer Nature Limited.

There has been a quest for superconductivity in the field of material science over the years. While a single-layered sheet of graphene is non-superconductive due to the absence of flat electronic bands as shown in figure 1(a), two sheets created by moiré pattern when placed vertically on top of each other and twisted just 1.1°, often referred to as 'magic angle', is proved to have exciting promises in this regard. The arrangement of carbon atoms in a twisted magic angle graphene generates a moiré pattern as shown in figure 1(b), resulting in a periodic superlattice structure. This altered super-lattice structure impacts the electronic band structure of graphene, leading to the emergence of flat electronic bands and the occurrence of correlated electron behaviour. This behaviour resembles the observed characteristics found in high-temperature superconductors. Due to the presence of inter-layer coupling between two graphene sheets, at a twist of about 1.1°, flat bands achieved at nearly zero Fermi energy results in either insulation at half-filling or zero-resistance state at up to 1.7 K critical temperature [35]. There has been experimental evidence supporting the presence of ferromagnetic behavior in magic-angle twisted BLG as well which is lacking in mono-layer graphene sheets [36, 37]. Such a ground-breaking discovery has further brought a

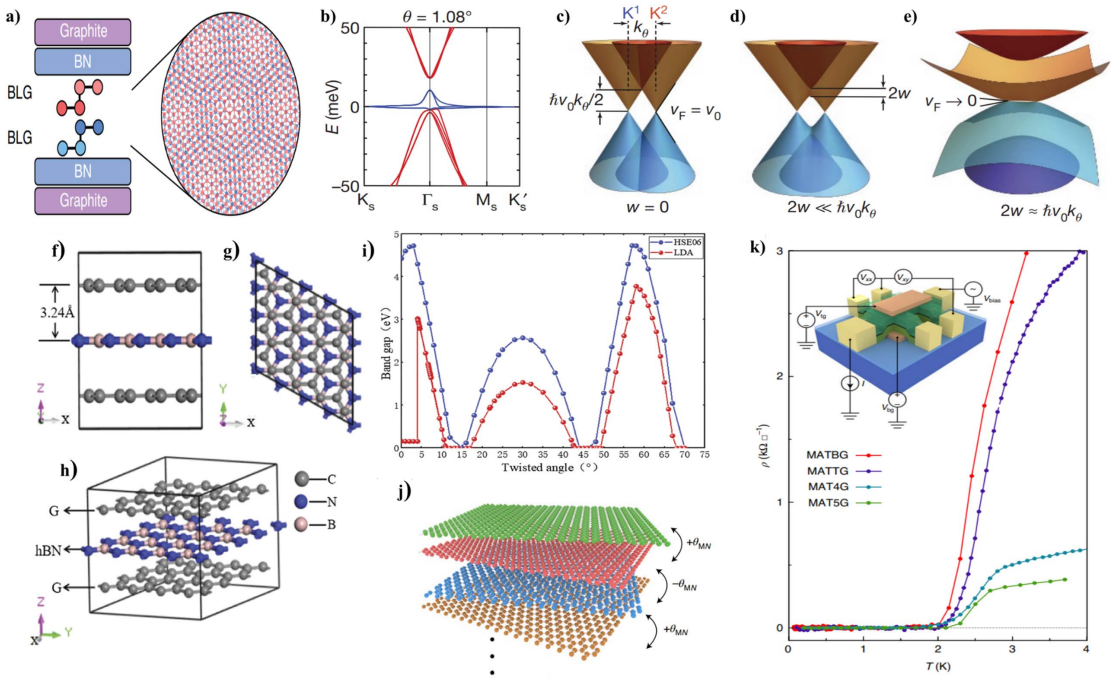

Figure 2. Overview of twist-dependent physical properties of Moiré superlattices consisting of 2D materials. (a) Schematic of twisted BLG sandwiched between two hBN sheets with graphite gates (heterostructure) [38]. Reproduced from [38], Copyright © 2020, The Author(s), under exclusive licence to Springer Nature Limited. (b) The band energy E at 1.08° magic angle twisted BLG; flat band is shown in blue colour [39]. (c)–(e) The energy band structure of twisted BLG, formed by the intersection of upper and lower layer Dirac cones [39]. Reproduced from [39], Copyright © 2018, Macmillan Publishers Limited, part of Springer Nature. All rights reserved. (f)–(h) Graphene/hBN/graphene heterojunction model (f) side view (g) top view (h) full three dimensional view [40]. (i) Band gap versus twist angle of graphene/hBN/graphene heterojunction [40]. Reproduced from [40]. © IOP Publishing Ltd. All rights reserved. (j) Twisted multilayer graphene with alternating twist angles. θ_{MN} is the magic angle specified for N-layer structure [32]. (k) Resistivity ρ versus Temperature T for two, three, four, five-layered magic angle graphene [32]. Reproduced from [32], Copyright © 2022, The Author(s), under exclusive licence to Springer Nature Limited.

tremendous impetus in the materials community over the last few years to explore different twist angles along with various stacking sequences of a range of 2D materials.

Unconventional superconductivity has often been an intriguing aspect of strongly correlated quantum materials where the interactions between electrons are so strong that they exhibit strange and exotic physical phenomena. Heavy-fermions [42], Quantum-spin liquids [43], High-T_c Superconductors [44] are one of few classes of the same. Though there have always been efforts to characterize these materials, the frameworks of proper solutions for analysing them theoretically is still limited. One novel approach to studying correlated quantum materials is using ultra-cold atoms by loading each atom at potential minima in optical lattices, resulting in the phase transition from Mott insulator to a superfluid [45] by varying interactions [46]. Bridging the length and energy scales between quantum materials and ultra-cold atom lattices, the twisted magic angle bilayer graphene (BLG) superlattice provides a new perspective to study strongly correlated materials (refer to figure 2(a)). The energy band structure of twisted BLG, with decreasing twist angle, formed by interacting Dirac cones is shown in figures 2(c)–(e) leading to flat bands as shown in figure 2(e) while band energy at magic angle is shown in figure 2(b) [39]. The twisting of layers has led to tuning the van Hove singularities in twisted BLG [47–52].

There exists a tremendous possibility of vertically combining different 2D materials along with their twisted configurations. The stacking of multiple layers in 2D materials has provided a space to engineer the properties even more than that of a single type of 2D materials [34]. Recent studies have been reported to capture the effects of twist angle between mono-layer graphene on a hexagonal boron nitride (hBN), but they have been found to showcase comparatively weak interlayer interaction due to large bandgap in hBN [53, 54]. A few groups [40, 55] have carried out first-principle studies on graphene-hBN based heterostructure to enhance and modify the electronic properties. Chen *et al* [40] studied graphene/hBN/graphene heterostructure (refer to figures 2(f)–(h)) by rotating upper layers of graphene and observed the Mulliken population, band gap, and charge density along with specific evolution regulars exhibiting novel electronic properties. Figure 2(i) shows the curve of band gap with respect to the rotation of upper layer graphene calculated by two different functionals. The twisting of layers has also led to the

showcase of the Hofstadter butterfly in graphene-hBN heterostructure [56, 57]. Further, the rotating angle has paved the way of modulating the optical responses of MoS_2 and $MoSe_2/WSe_2$ heterostructures [58]. Recently, Superconductivity has been found in tri-layer twisted magic angle graphene [59, 60]. Though they share similarities with magic angle twisted BLG, the response to external magnetic and electric fields is different. Park *et al* [32, 61] extended these studies to four and five layers (refer to figure 2(j)) to see whether they can classify these superconductivity phenomena into some sort of moiré superlattice superconductors and found superconductivity through same flat bands. But they observed variations as well, predominantly between two and more than two layered systems. Figure 2(k) shows resistivity ρ versus temperature T for two, three, four, five-layered magic angle graphene sheets.

The brief discussions on twisted bi-layer graphene and other multi-layer heterostructures, as presented above, clearly show that there has been an increasing interest in such engineered nanostructures over the last few years due to their unprecedented multi-physical properties. The research is so fascinating and rapidly evolving that it has led to a new field, called Twistronics. Thus there is a strong rationale to abridge outcomes of the relevant literature concerning twist-dependent properties of BLG and other multi-layered 2D heterostructures. In this review paper, we would analyse the recent developments on twisted bilayer and multi-layer nanostructures formed out of single or multiple 2D materials, and subsequently highlight their broad-spectrum potential to meet the functional demands of modern engineering applications.

2. Outline and organization

The broad scope of this paper is highlighted in figure 1 including a list for potential 2D materials that can constitute the twisted nano-heterostructure. After providing an introductory overview in section 1, we have discussed the twisted architectures of bi-layer and multi-layer graphene along with other 2D materials in section 3. The fabrication techniques of such complex nanostructures are also briefly discussed in this section. In the following three sections, we have discussed different twist-dependent physical properties systematically, such as (1) electronic properties and superconductivity, (2) optical, magnetic and other multi-physical properties, (3) mechanical properties and their correlations with different physical properties. The next section deals with evolving trends and future research directions, such as the exploration of the multi-objective design space of stacking sequence and interlayer twisting (including exploitation of the recent advances in artificial intelligence and machine learning), possibilities of expanding the design space through nano-scale architectures like layer-wise multi-directional translation, defect engineering and strain engineering, critical issues of interfacial effects and stability, integration with other materials and formation of nanocomposites, manufacturing complexity, scalability and service-life effects. Finally, a summary of the review and concluding remarks are presented.

3. BLG superlattices and multi-layer heterostructures with twisted configurations

The tuning of physical properties by varying the relative angle about a perpendicular axis to the plane of different layers of 2D materials leads to a novel research field of Twistronics. Owning to relative interlayer twisting, a moiré superlattice is formed as shown in figure 3(c). The electronic band structure of perfect mono-layered graphene has conical forms, also referred to as Dirac cones which touch at K and $K\prime$ points (the specific region of reciprocal-space which is closest to the origin) at the Fermi level in the hexagonal Brillouin zone as shown in figures 3(a) and (b) [62]. When two separate Brillouin zones of twisted bi-layer graphene come together, there happens a shift of Dirac cones by ΔK, reconstructing the electronic band structure as shown in figure 3(e). The mini Brillouin zone formed from the difference in two K (or $K\prime$) wave vectors of the two graphene layers is shown in figure 3(d). With twisting of bi-layer graphene, their Brillouin zones too rotate, ending with a certain angle between them. The energy bands at overlapping region bend resulting in van Hove singularity [63, 64], enhancing the optical absorption [65, 66] of twisted BLG. Using local density of states (LDOS), the presence of van Hove singularity is also confirmed by varying the twisting angle from 1° to 10° [67].

When the twist angle is higher, then with the reduction of it, the Fermi velocity starts behaving sensitively and periodicity keeps changing [68]. Particularly at lower twisting angles, it shows high-periodicity and uniformity of electrons system showcasing vivid electronic properties [69]. When this twisting angle is around 1°, the van Hove singularities in the two electronic bands are close enough, forming a nearly flat band in 'magic angle' twisted BLG [70] that leads to superconductivity. When two graphene sheets align in this way, it results in a restructuring of the atoms into domains where electrons get trapped in a localized space. But as natural electrons tend to repel each other, so to limit this strong interaction, electrons align their spins resulting in magnetic properties or result in insulators/superconductors. Researchers have shown

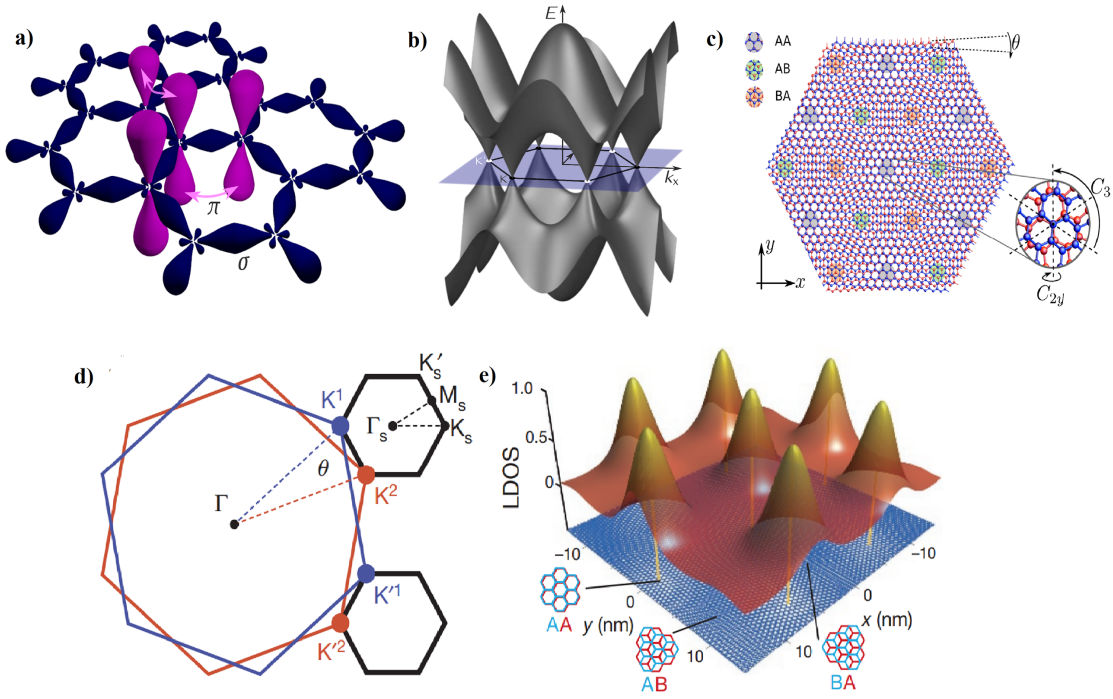

Figure 3. Electronic band structure of pristine graphene and magic angle twisted BLG. (a) A single layer graphene sheet showing sigma and pi bonds. (b) Electronic structure of pristine graphene. (c) Twisted graphene by an angle θ in twisted BLG moiré superlattice [41]. Reprinted figure with permission from [41], Copyright (2018) by the American Physical Society. (d) Mini Brillouin zone formed from the difference of two K (or $K^{'}$) wave vectors for two graphene layers [39]. Reproduced from [39], Copyright © 2018, Macmillan Publishers Limited, part of Springer Nature. All rights reserved. (g) Normalized LDOS (local density of states) calculated for flat bands with $E > 0$ at 1.08°. We notice strong concentration of electrons in the regions having AA stacking and diminished in BA and AB stacking [39].

the presence of phonons (atomic particles responsible for vibrations in solid materials) in the vicinity as well [71]. It's quite interesting for the same material to have superconductivity and magnetism, providing exciting new possibilities for engineering applications.

Physical realization and experimental validation of the computationally identified properties are crucial along with further experimental innovations. Twisted BLG is fabricated by stacking a layer of graphene on top of the other forming a Lego-like structure. Separation of a single layer of graphene from it is bulk graphite crystalline form is quite a cumbersome process, hampering its many potential applications. Though various synthesis methods are there such as chemical vapour deposition (CVD) [72, 73], thermal-deposition of silicon carbide [74, 75], chemical reduction of exfoliated graphene oxide [76, 77], each method has its pros and cons. Twisted bi-layer graphene has been folded from single-layered graphene [65, 78–81] and conventionally prepared using chemical vapour deposition to stack two single-layered graphenes [82–84]. Similar is the case of transfer alignment in which a monolayer graphene is first grown on a metal catalyst using CVD and the monolayer graphene is then transferred onto a substrate. A second monolayer graphene is grown on a separate substrate and then by aligning and stacking the two graphene layers with the desired twist angle, twisted BLG is obtained [85]. Another method is the process of electrochemical intercalation which involves the insertion of foreign atoms or molecules between the layers of graphene to create a twisted BLG structure. This method relies on the intercalation of small molecules or ions into a graphene lattice, followed by thermal or chemical treatments to induce a twist [86]. In alignment with the earlier method of fabricating twisted BLG, chemical functionalization involves modifying one or both layers of graphene with functional groups or molecules, leading to a change in the interlayer coupling and resulting in a twisted structure [87]. Some other methods include Joule heating and then rapid cooling of polyaromatic hydrocarbons on Nickel [88], controlled folding of single-layered graphene [89], chemical vapour deposition by decaborane-coated Cu foil [90], and successive transfer of single-layered graphene [91]. A group of researchers [35] made their twisted bi-layer graphene magic angle device by stacking vertically single-layered graphene using the mechanical transfer method to study superconductivity. Another common method to fabricate twisting-based electronic devices is the tear-and-stack technique [91, 92]. For fabrication of other twisted nano-heterostructures, one of the reasonable paths is to fabricate high-quality 2D layers using suitable fabrication techniques, and later on, stack the layers by repeat transferring and stacking.

Post the remarkable findings on the properties of magic-angle twisted BLG and its successful fabrication, keen explorations have been made to apply this twist science to other 2D van der Waals superlattice heterostructures [93–97] for the quest of achieving exciting properties [98–101]. Multi-Layered 2D superlattices may possess more fascinating properties due presence of multiple layers [59, 102] and stacking sequences viz., AA or AB stacking of layers [103]. In AA stacking, the carbon atoms in two layers have same lateral coordinates while in AB stacking, one layer of graphene is relatively shifted with respect to other one by the edge length of the hexagon lattice [104]. To leverage the advantage of unusual properties, a large number of moiré 2D materials is being created using hBN, graphene, transition metal dichalcogenides, or magnetic semiconductor crystals [105]. These 2D materials can be classified as Homo-bilayers (which are produced when two layers of the same 2D materials are placed vertically with some relative twisting), Hetero-bilayers (which are produced when two layers of different 2D materials are stacked vertically with some relative twisting) and Multi-layers (when multiple layers of 2D materials are stacked vertically with some relative twisting) [106]. As a result, The methodologies for designing and fabricating moiré 2D heterostructures also vary, and needless to say the research in this domain is still at the nascent stage.

While the field of Twistronics started with a focus on the electronic properties of twisted BLG, other multi-physical aspects have subsequently been investigated including the possibility of creating different twisted 2D structures using a large number of available 2D materials. In the subsequent sections, we would concentrate on twisted bi-layer and multi-layer nano-structures (with 2D layers of same and different materials) focusing on a range of critical properties such as electronic, optical, mechanical and other multi-physical aspects.

4. Twist-dependent electronic properties and superconductivity

The twisting of angle between the two layers of graphene and the inter-coupling interaction between them defines the shape of the electronic band structure. Talking of pristine monolayer graphene, researchers long speculated it to have correlated states (CSs), marked by collective instead of individual charge carriers. Superconductivity and Mott insulation are likely to take place in materials with collective electrons sharing the same energy. Such states exist in flat bands in the premise of a saddle point [107] i.e. in momentum space, saddle points are locations where the energy band experiences energy maxima and minima along orthogonal lines. In 2D materials, van Hove singularities are amplified density of state peaks produced by saddle points that can be easily recognizable by scanning tunneling spectroscopy [63]. In Pristine monolayer graphene, this saddle point lies at some electron volts higher than that of Fermi level, and with just an applied voltage, it is not feasible to raise the Fermi level to the saddle point.

The twisting in BLG by moiré pattern tends to bring down the saddle points. At large twist angles the Dirac cones of the two graphene layers are far and tunnel coupling to the adjacent layer has no effect on the low energy states in that layer. The gap between the valance and conduction band van Hove singularities is equal to double the isolated layer energies at the Dirac cone intersecting points [108] (more like behaving as two weakly coupled layers). When the angle of twist is reduced, the hybridization between the layers grows significantly, and the van Hove singularities of the two layers come closer. At around 1.1° firstly insulating behaviour below $4K$ is observed, and with further decreasing temperature and with varying charge carriers leads to superconducting states (SC) [35]. A typical schematic of a twisted bi-layer graphene device is shown in figure 4(a)(i). The longitudinal resistance against temperature and charge density showing phases as band insulator (BI), SC state, metal and band CS are shown in figure 4(b) [109]. The resistance measured in two devices $M1$ and $M2$, with twist angles of 1.16° and 1.05° respectively is shown in figure 4(d) indicating nearly zero resistance in $M2$ at very low temperatures [110].

These findings have opened a new way of looking at correlated physics for high-temperature superconductors. Their similarity with the magic angle phenomena has led to an understanding of the physics of high-temperature superconductors [111, 112]. In fact, twisted bi-layer graphene has the advantage of doping it by electrostatic gating to improve the carrier density in an electronic device while doping high-temperature superconductors requires chemical synthesis. Lu *et al* [109] fabricated a more refined magic angle twisted graphene-based device by employing a mechanical cleaning process to get rid of air bubbles, strains, or impurities, and at around -2 moiré band filling factor, observed superconductivity below the critical temperature of $3K$, a higher temperature compared to [35]. They also observed insulating states at moiré band filling factors of 0 and ± 1, as shown in figure 4(c).

The formation of bandgap in twisted graphene-based superlattice due to the creation of Brillouin zone folding which in fact is the result of variations present in the two wave vectors of the two graphene sheets has potential properties still to be fully leveraged for incorporation into new electronic devices. Codecido *et al* [113] have shown in their work, both insulating and SC states at about 0.93° which is kind of less compared to earlier findings at about $1.1 \pm 0.1°$, revealing the 'magic angle' range of twisted graphene to be broader

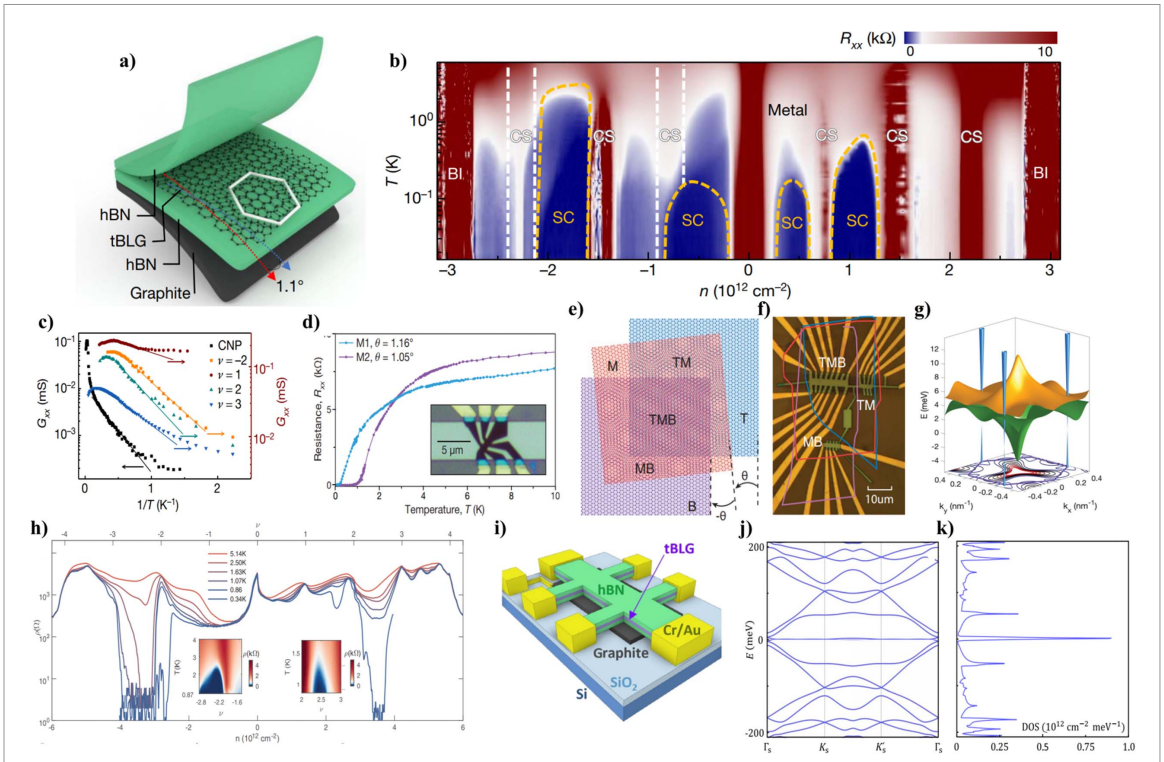

Figure 4. Superconductivity in twisted graphene superlattices. (a) Schematic of a typical twisted BLG device [109]. (b) Longitudinal resistance against temperature and charge density showing phases as band insulator (BI), superconducting state (SC), metal and band correlated state (CS) [109]. (c) Conductance G_{xx} vs inverse temperature at carrier densities corresponding to moiré band-filling factor, ν, at $0, 1, \pm 2$ and 3 [109]. Reproduced from [109], Copyright © 2019, The Author(s), under exclusive licence to Springer Nature Limited. (d) Resistance measured in two devices $M1$ and $M2$, with twist angles of 1.16° and 1.05° respectively [110]. Reproduced from [110], Copyright © 2018, Macmillan Publishers Limited, part of Springer Nature. All rights reserved. (e) Schematic of twisted tri-layer graphene with θ and $-\theta$ alternate twist angles. The middle layer is twisted by θ with respect to the top and bottom layers, which are aligned to each other [59]. (f) Optical microscope image of twisted tri-layer graphene device fabricated in the three-layer overlapped region. Two twisted BLG devices fabricated in the top and middle layers, and middle and bottom layers [59]. (g) Band structure of magic-angle twisted tri-layer graphene plotted on the mini Brillouin zone, as purple marked in the bottom face. The orange and green colours represent conductance and valance bands [59]. (h) Resistivity, ρ, as a function of moiré band-filling factor, ν, at different temperature values in twisted tri-layer graphene. From [59]. Reprinted with permission from AAAS. The superconducting regions is seen at $\nu < -2$ and $\nu > 2$ [59]. (i) Schematic of twisted BLG device observing superconductivity below magic angle [113]. (j), (k) Moiré electronic band structure calculation of 0.93° twisted BLG for energy dispersion and density of states (DOS) [113]. From [113]. Reprinted with permission from AAAS.

than previously expected (refer to figure 4(i)). This finding is indicated by the calculated moiré band and density of states of 0.93° in twisted bi-layer graphene (refer to figures 4(j)–(k)).

Moiré patterns can largely alter electronic properties [63]. The occurrence of localized flat bands nearby Fermi level at the magic angle gives rise to many-body effects [71, 116, 117] resulting in charge carriers lacking enough kinetic energy to escape the strong interactions among themselves. This leads to strongly CSs, and when there are strong interactions among particles, exotic properties are all over at display. The gap of the rotated Brillouin zone separates the layers in twisted graphene and so a significant change in momentum is necessary for an electron to tunnel between them. But at small twist angles, interlayer tunneling immensely hybridizes the layers leading to modification of band structure and at 1.1° twist angle the Fermi velocity goes fading and a low-energy band appears [118, 119].

Yu *et al* discussed interlayer contact conductance in twisted graphene layers synthesized by chemical vapour deposition method [114] and found the interlayer contact conductance at a small twist angle to be higher in AB stacked configuration. Figure 5(a) shows SEM (scanning electron microscopy) images of as-grown graphene/copper samples. The normalized current distribution of certain regions of AB stacked configuration with varying twist angles is shown in figure 5(b) and the interlayer conductance is shown in figure 5(c). Polshyn *et al* [115] reported electrical transport measurements of twisted BLG devices having twist angles varying from 0.75° to 2° up to room temperature and observed resistivity, ρ, to vary linearly with the temperature over large temperature ranges as shown in figure 5(d). Choi *et al* [71] studied the electron-phonon interaction in twisted BLG using a tight binding approach in the case of electrons and atomic force constants for phonons and found the coupling strength to be nearly comparable to the electron density of states and it became greater than 1 near half-filling energies of the flat bands. They showed the effects of lattice relaxation on the electronic structure of magic-angle twisted graphenes, as shown in

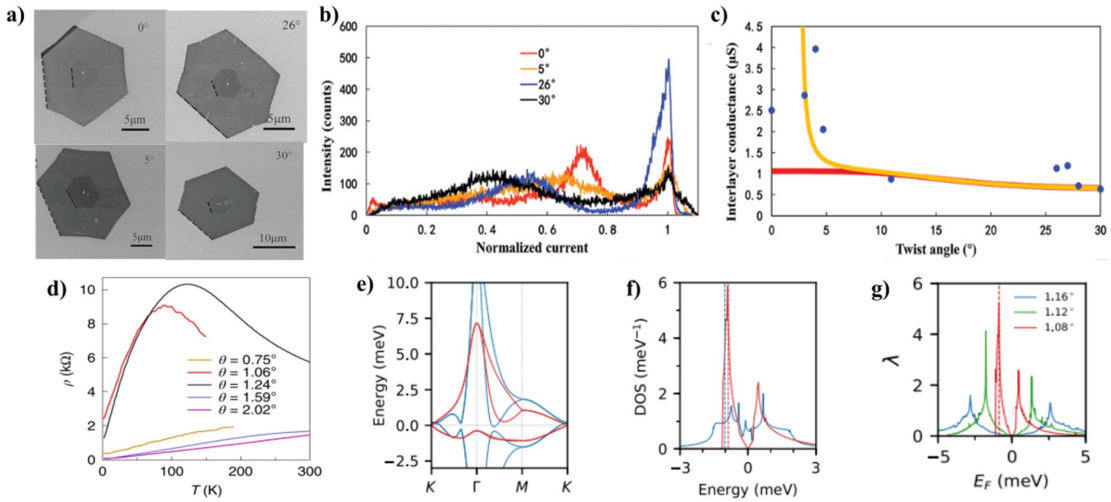

Figure 5. Electronics of twisted BLG. (a) SEM image of twisted BLG regions from 0° to 30° [114]. (b) Normalized current distribution of certain regions of AB stacked twisted BLG with different twist angles [114]. (c) Inter-layer conductance of twisted BLG at various twist angles. The yellow and red curves show a phonon-assisted transport mechanism with and without considering the re-normalization of the Fermi velocity respectively [114]. [114] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Resistivity ρ as a function of temperature T measured in devices with different twist angles [115]. Reproduced from [115], Copyright © 2019, The Author(s), under exclusive licence to Springer Nature Limited. (e), (f) Electronic structure of magic angle twisted BLG. Tight-binding (e) band structure (f) density of states per spin per moiré supercell for relaxed (red) and non-relaxed (black) structures at 1.08° [71]. (g) Total phonon-electron coupling strength λ in twisted BLG with respect to Fermi energy E_F [71]. Reprinted figure with permission from [71], Copyright (2018) by the American Physical Society.

figures 5(e) and (f). Total phonon-electron coupling strength λ in twisted bi-layer graphene with respect to Fermi energy is shown in figure 5(g).

In addition to twisted BLG, researchers have explored multilayer twisted graphene. Hao *et al* [59] studied electric-field tuned superconductivity in alternating-twist tri-layer graphene, as shown in figures 4(e) and (f) the band structure of which is further shown in figure 4(g). The resistivity as a function of moiré band-filling factor, ν, at different temperature values for twisted tri-layer graphene, revealing SC regions at $\nu < -2$ and $\nu > 2$ is shown in figure 4(h). Klein *et al* [120] recently created a device by layering magic angle bernal BLG between two offset layers of insulating hBN, one layer of hBN was placed above the entire structure, aligned with the top layer of graphene while the second layer of hBN was stacked offset by 30° with respect to the top layer. A wide potential of tunable and switch-able SC electronics is possible owning to the sandwich structure's special alignment, which allows bi-stable switching between correlated insulating, SC, and metallic states using an electric field or gate voltage. Lee *et al* [121] studied a system of twisted two bernal stacked double bi-layer graphenes, twisted relative to one another and observed a lower symmetry as compared to twisted bi-layer graphene. Absence of C_2 rotation symmetry leads to removal of band touching at the Dirac points and further directing to a low energy effective description requiring one instead of two narrow bands per spin and valley. Due to voluntary symmetry breaking and strong electronic correlations, the 2D moiré heterostructures i.e. 1T − MoS_2, GeSe, In_2Se_3 and 1T − $MoTe_2$ have shown unconventional ferroelectric responses as observed in some theoretical as well as experimental findings [122, 123]. With an edge of multiple stacking options and more number of layers leading to more tunable possibilities [124–126], some of them are recently synthesized and an enhanced focus has been placed on physical realization of these complex nanostructural configurations [60, 127].

5. Twist-dependent optical, magnetic and other multi-physical phenomena

The properties of twisted BLG are largely dependent on the twist angle between the two stacked bilayers [115, 128–131]. As discussed in the preceding sections, at around magic angle 1.1°, the emergence of flat band leads to strong correlation occurrences viz., superconductivity [110], Mott insulator, ferromagnetism [37] topological Chern insulator [132] and quantized anomalous Hall effects [133]. The energy difference between the two van Hove singularities in the two bands must match with that of the excitation photon energy that leads to the resonant absorption [134, 135] and the exotic spectral features [136] at this specific twist angle. The optical-based studies have emerged as an important tool in studying structures in graphene [137, 138]. Raman Spectroscopy (a spectroscopic technique typically used to find the vibrational and the

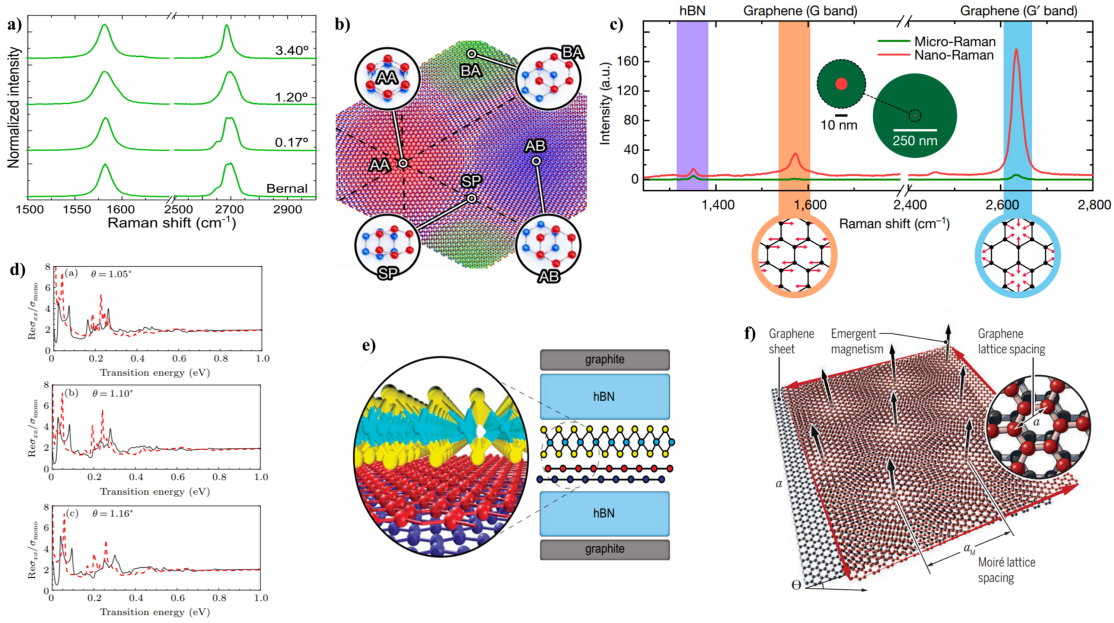

Figure 6. Optical and magnetic phenomena of twisted BLG. (a) 2D and G band Raman spectra of a Bernal bilayer graphene [141]. Reproduced from [141]. © IOP Publishing Ltd. All rights reserved. (b) Schematics of neighbouring BA and AB stacked domains, topological points (AA) and strain solitons (SP) [142]. (c) Nano-Raman and Micro-Raman spectra. A Nano-Raman spectroscope is capable of resolving optical information below diffraction of light [142]. Reproduced from [142], Copyright © 2021, The Author(s), under exclusive licence to Springer Nature Limited. (d) The conductivity spectrum of unrelaxed (red) and relaxed (black) twisted BLG at 1.05°, 1.10° and 1.16° [143]. Reproduced from [143]. © 2021 Chinese Physical Society and IOP Publishing Ltd. (e) A twisted bi-layer graphene/WSe_2 heterostructure, encapsulated with hBN and graphite [144]. From [144]. Reprinted with permission from AAAS. (f) The moiré pattern making the twisted BLG both having conducting edges, with electrically insulated bulk and magnetic [36]. From [36]. Reprinted with permission from AAAS.

rotational modes of molecules) is used widely in the case of graphene to study optical and electronic phenomena employing non-destructive testing of the structure [139, 140]. Barbosa *et al* [141] studied the Raman spectra of twisted BLG corresponding to their twisting angles ranging from 0.03° to 3.40°, where the twist angles between the bi-layers are determined by analysing the associated moiré lattices, imaged by scanning impedance microscopy. They used three different wavelengths of excitation laser lines considering the main Raman active graphene bands namely 2D and G. The study revealed that electron-photon interaction influences the G band's linewidth near the magic angle. 2D and G band Raman spectra of a Bernal bilayered graphene is shown in figure 6(a).

The 2D band shape having twist angle less than 1° is governed by crystal lattice depending upon stacking sequence (AB and AA) of bi-layers and strain soliton regions [142]. The Schematics of neighbouring BA and AB stacked domains, topological points (AA), and strain solitons (SP) are shown in figure 6(b). The difference between the nano-Raman and micro-Raman spectra is shown in figure 6(c). Twisted double bi-layer graphene was found to host a topological gapped phase when half of the flat bands are filled, having Chern number equal to 2 at charge neutrality [145, 146]. Wang *et al* [147] studied the edge and bulk phenomena of double bi-layer twisted graphene using transport and chemical potential measurements combined with theoretical calculations and showed the twisted double bi-layer graphene to have metallic edge transport while being insulated in bulk at the same time.

Optical spectrum can reveal a lot about electronic band structure, quasiparticles [148], and many-body interactions [149] in graphene-based systems. For studying optical absorption properties Moon *et al* [150] showed the importance of spectroscopic characteristics in identifying the rotation angle between two layers. Wen *et al* [143] studied the optical conductivity of twisted BLG near magic angle taking into consideration the effects of lattice relaxation (lattice relaxation tends to modify flat bands leading to shifts of peaks in optical conductivity). They showed the optical conductivity spectrum is distinguished by a series of characteristics peaks linked to van Hove singularities in the electronic band structure and the peak energies develop systematically with the twisting angle. The conductivity spectrum of unrelaxed (red) and relaxed (black) twisted graphene structures at 1.05°, 1.10° and 1.16° is shown in figure 6(d)

Transition metal dichalcogenides such as MoS_2, WSe_2, $MoSe_2$ and WS_2 are excellent for strong light-matter interactions in optoelectronic devices because of the van Hove singularity-induced significant optical absorption and exciton production in the visible region of the electromagnetic spectrum. Like most

of van der Waals heterostructures they posses weak van der Waals interaction and strong covalent interactions [151–153].

Recently by inducing spin–orbit coupling, magic angle twisted BLG is shown to behave as ferromagnets [36, 144]. The strong correlation among electrons within the flat bands stabilizes the correlated insulating states at the half and quarter fillings and with spin-orbital coupling transforms Mott insulators into ferromagnets. The effect of spin–orbit coupling is investigated by using transport measurement to study the properties of moiré band and its related quantum phases of twisted bi-layer graphene/WSe_2 heterostructure figure 6(e). The twisting of a layer by a small angle, θ, makes the twisted BLG to have conducting edges, with electrically insulated and magnetic, as shown in figure 6(f) [36]. Xu *et al* [157] studied a Hubbard model for superconductivity in twisted multi-layered graphene and showed that in contrast to twisted bi-layer graphene which invokes ferromagnetism [158] the twisted multi-layer graphene relied on effective anti-ferromagnetic interaction.

6. Twist-dependent mechanical properties and correlations

Besides investigating the electronic, optical and magnetic properties of twisted graphene and other 2D materials, the mechanical properties have also been explored for twist-dependent programming, characterization and correlation with other physical properties. Researchers have shown the dependence of heterostrain (an unequal strain on the two monolayers of graphene sheet) and uni-axial strain on the moiré pattern and flat band based optical and electronic properties [159–163]. Using Scanning Tunnelling Microscope Mesple *et al* [160] showed the dependence of flat band formation on the relative deformation among the layers. The weak van der Walls interlayer interaction often allows the stacking sequence and a way of straining the heterostructures and in the case of twisted graphene layers the divergence in the superlattice. Huder *et al* strained the two layers of graphene and showed the suppression of Dirac cones leading to flat bands [154] and that the energy spectrum of twisted bi-layer graphene is more severely varied in the case of heterostrain than when layers are homostrained, altering the band structure and providing more programmability [164]. The effect of uniaxial strain on moiré structure of twisted graphene is shown in figures 7(a)–(c). The local density of states in *AB* and *AA* regions with effects of heterostrain and homostrain are shown in figures 7(d)–(f). Using scanning tunnelling spectroscopy and scanning tunnelling microscopy researchers [165] studied the effects of heterostrain on a number of twisted graphene bilayer configurations near the first magic angle and found the heterostrain to largely manipulate the atomic alignment leading to change in geometric structure and overall physics of BLG and $\alpha - Mo_2C$ heterostructure.

Carr *et al* [155] reported compression dependence studies on twisted graphene nanostructures using *ab initio*, a first-principle approach combining density function theory along with maximally localized Wannier functions [166] and showed the role of relaxation on low energy states of twisted bi-layer graphene. Compression leads to producing correlated behaviour marked by the presence of flat bands at twist angles which increase with increasing compression. At 5% compression for twisting of 1.47° and at 10% compression for twisting of 2°, the flat band regime is achieved figure 7(g). The shift in momentum space in the two Dirac cones results from the twisting of layers and by interlayer coupling strength between them, as shown in figure 7(h). Nguyen *et al* [167] reported a comprehensive study on the electronic properties of twisted multilayer graphene using atomistic calculations and presented the possibilities for tuning the electronic properties using external fields and by applying strains/vertical pressure.

From a different perspective, for successful utilization of the unprecedented properties of twisted bilayer and multilayer 2D superlattices in nano-scale devices, it is of utmost importance to study the mechanical properties. A significant number of recent studies have investigated multilayer 2D materials for their mechanical characterization [168–171]. Few research groups have started exploring similar directions for their twisted configurations recently. Mukhopadhyay *et al* [156] studied elastic moduli of multi-layered twisted heterostructures based on an effective mechanics-based analytical approach. They presented a generic framework, applicable from magic angle twisted bi-layer graphene to some random twist angles and further to any multi-layered heterostructure with any possibilities of rotation angles between layers (a typical three-layered heterostructure is shown in figure 7(i), where each of the layers can have an arbitrary twist). Young's moduli (E_1 and E_2) of two-layers configurations by rotation of one layer are shown in figures 7(j) and (k). Chandra *et al* studied the buckling behaviour of twisted 2D materials and heterostructures with moire patterns [172]. Mechanical properties (including strength, stiffness and elastic modulus) of twisted BLG along with grain boundaries are reported by Zhang and Zhao [173]. Though there has been studies on mechanical properties of 2D single-layer materials, bilayer twisted graphene configurations and 2D multilayer heterostructures by stacking without taking into account the considerations of twisting between layers [168, 174–179], the exploration of elastic properties and mechanical strength of twisted multi-layer 2D materials and heterostructures are still limited in the literature. Most importantly, the possibility of

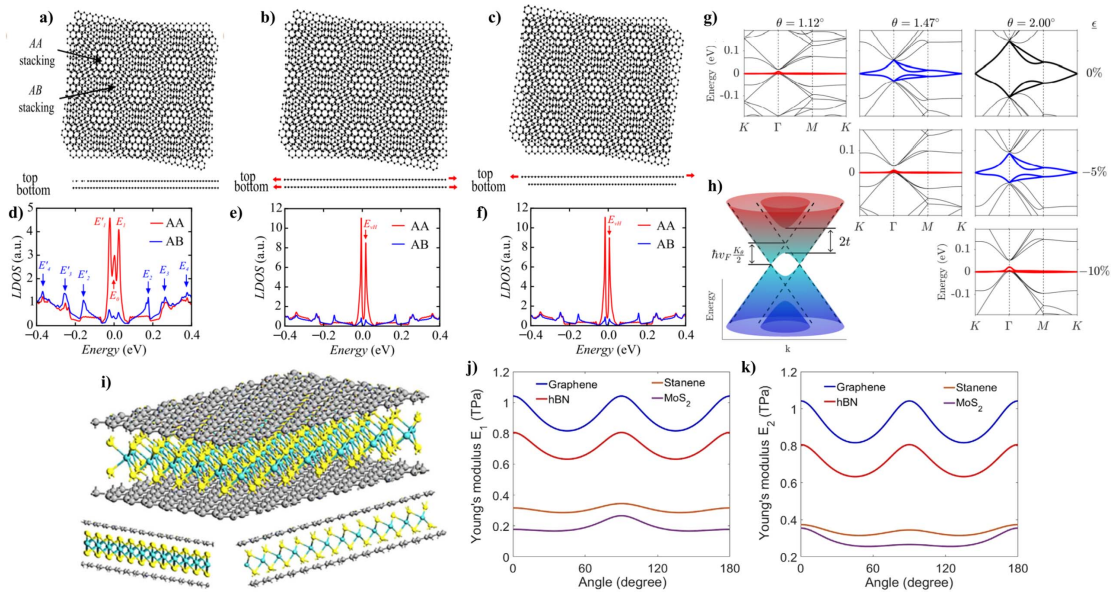

Figure 7. Mechanical characterization of twisted BLG and other heterostructures. (a)–(c) Strained twisted BLG (a) twisted BLG without strain (b) twisted BLG with 10% of uni-axial strain applied to the two layers. (c) twisted BLG with 10% of uni-axial strain applied to top layer only [154]. (d)–(f) Local DOS in *AA* and *AB* regions (d) corresponding to a structure with 0.35% heterostrain (e) corresponding to a unstrained structure (f) corresponding to a structure with homostrain of 0.35% [154]. Reprinted figure with permission from [154], Copyright (2018) by the American Physical Society. (g) Band structures for twisted BLG under compression, ϵ, from the *ab initio* tight-binding model [155]. (h) The shift in momentum space due to twisting in the two Dirac cones with interlayer coupling strength λ_0 [155]. Reprinted figure with permission from [155], Copyright (2018) by the American Physical Society. (i) A three-layer heterostructure (3D view and side views) made of two different 2D materials [156]. (j), (k) Effective Young's modulus E_1 and E_2 of a bi-layer nanostructure with rotation of one layer [156]. Reproduced from [156]. CC BY 4.0.

simultaneously achieving multiple mechanical behaviour (such as multi-directional strength and stiffness, direction-dependent vibration and wave propagation modulation, controlling buckling under complex multi-directional and multi-modal loading conditions, simultaneous programming of mechanical and other multi-physical properties) is yet to be investigated through nano-engineered twisting of multi-layered 2D materials.

7. Evolving trends and future roadmap

With immense multi-physical properties under the hood, stacked 2D heterostructures particularly magic-angle graphene and other twisted configurations of multi-layer 2D materials are being continuously explored for further new findings. Considering the current momentum of research in this direction, it would be compelling to explore a range of new possibilities for unveiling exciting physical properties. In this section, we provide an overview of the prospective future research trajectory and evolving trends concerning twisted 2D nanostructures along with some of the critical areas that need immediate attention from the scientific community.

7.1. Coupled multi-objective design space of stacking sequence and interlayer twisting

The aspect of stacking the layers of similar or different 2D materials on top of each other results in variation in properties, and coupling them with interlayer twisting in a multi-layered 2D heterostructure system could lead to alluring exotic optical, electronic and magnetic phenomena. The vast possibility of involving a range of 2D materials, their number, stacking sequence and individual twisting angles is far from being explored. Moreover, the aspect of multifunctional design by involving multi-objective goal attainment algorithms with application-specific weight assignments to different individual goals of optical, magnetic, mechanical or electronic performance is still uncharted territory. The possibility of weighted multi-objective target space allows the designers and scientists to infuse the notion of engineering judgment while developing bespoke 2D heterostructures. These controllable degrees of freedom could lead to multi-physical tailorable and tunable properties of unprecedented extent, which would certainly drive the interests of the researchers for the foreseeable future. The only concern though would be the stability of the nanostructures during synthesis with so many parameters to control that needs careful consideration. This indicates that a combined

computational and experimental framework needs to be adopted for exploring different possible configurations of twisted nano-heterostructures in the near future.

7.2. Layer-wise defect and strain engineering coupled with twist

Imperfection is a part of nature; in one way or the other, defects are inevitable in the synthesis of 2D nanomaterials. It would be interesting to explore the variation in physical properties (electronic, optical, magnetic, mechanical and other multiphysical features) by intentionally imparting defects in the layers of 2D materials (such as vacancy, substitution, grain boundary, stacking faults etc), to investigate the correlation between defects and properties along with twist angle in multi-layered heterostructures. Note that different forms of strains, which can normally be regarded as irregularities, can also be deliberately introduced within different twisted layers in a programmed way to modulate the physical properties along with the above-mentioned design space.

7.3. Translation with twist

Layer-wise twist in bi-layer and multi-layer 2D materials essentially provides more design variables for modulating the effective physical properties of the resulting nanostructures. Following a similar line of research, along with twist, there are further possibilities of adding new sensitive features at each layer, which are discussed in this subsection. Owning to the weak interlayer coupling between the layers of 2D heterostructures, it allows the twisting and stretching of layers with respect to each other. It would be interesting to explore their individual and coupled behaviour for tuning a range of electronic, optical and magnetic properties for single or multi-objective goals with fascinating programmable features.

7.4. Interfacial effects and stability

Interfacial effects between the layers in bi/multi-layer twisted graphene and other nano-heterostructures play a crucial role in determining their properties, but these effects are difficult to quantify and predict. More investigations including experimental validations are essential in this direction.

Another crucial challenge in the physical development and realization of different computationally predicted twisted configurations is the aspect of stability. These complex bi/multi-layered nano(hetero)structural configurations are susceptible to degradation under various environmental conditions (both during manufacturing as well as during service condition) which needs comprehensive investigations before real-life and industry-scale implementation.

7.5. Integration with other materials and formation of nanocomposites

For a range of technologically demanding applications, nanocomposites are formed along with the 2D material nanostructures. The integration of bilayer twisted graphene and different engineered nano-heterostructures with other materials and devices is challenging, as it requires the development of new computational and experimental techniques for their design, synthesis and characterization. Comprehensive research in such direction is still almost non-existent, and needs further attention for various engineering applications.

7.6. Exploitation of machine learning and artificial intelligence

With tremendous recent advances in the field of machine learning and artificial intelligence, the materials science community has grown interests to integrate the behaviour of materials with data-driven approaches for achieving computational efficiency as well as exploring hitherto unexplored design domains. The field of 2D materials with the vast scope of combining their different variants with numerous possibilities of stacking sequences along with design parameters like twisting angle, strains, introduced defects and translations would embrace such data-driven approaches in the near future for identifying target combinations of the parameter space that would be impossible to explore solely using conventional methods of simulations and experiments. Besides efficient prediction, the emerging machine learning and artificial intelligence algorithms would be useful in feature identification, sensitivity analysis, optimum design for multi-objective goals and uncertainty quantification. However, the researchers need to be considerate about the accuracy of the developed machine-learning models to avoid any misleading outcomes including the emerging concepts of responsible artificial intelligence.

7.7. Exploring prospective applications in critical technologically demanding sectors

With the advances and possibility of a wide range of rich physical properties, prospective applications and product development needs more attention in the areas of twisted BLG and other heterostructures. Such advanced engineered nanostructures can play crucial roles in the areas of spintronics and quantum computing. These materials have been found to exhibit strong absorption and large nonlinear response,

which make them promising candidates for applications in photodetection, light harvesting and photovoltaics, including applications in solar cells. A crtical area of application for twisted nano-heterostructures which has not been adequately explored is energy storage, including the use of graphene-based materials as supercapacitors and batteries. Due to their high electrical conductivity and large specific surface area, these materials have shown promise for such applications. In recent years, researchers have made significant progress in developing high-performance energy storage devices based on 2D twisted heterostructures, such as graphene/boron nitride heterostructures for supercapacitors. There exists a vast scope for improving the energy storage performance by identifying optimal heterostructural configurations including the aspect of layer-wise twist.

A range of 2D materials have been found to exhibit high mechanical strength and toughness, which makes them potentially useful for various mechanical applications, such as flexible and stretchable electronics. Most importantly, the aspect of layer-wise twist would bring the advantage of dealing with multi-directional stresses using a single heterostructure. Extremely promising mechanical properties of these engineered materials also make them suitable for prospective applications as building blocks in the construction of novel composites and advanced nanodevices that would essentially lead to creation of nano-scale metamaterials [180–183].

7.8. Complexity in fabrication techniques and scalability

Due to the engineering at a nano-length scale and being atomically thin, it is quite complicated to fabricate single and multi-layer 2D materials and their heterostructures. Moreover, providing the accurate amount of layer-wise twists along with other proposed design-level parameters like programmed defects, direction-dependent strain, and translation would demand a high level of precision in the fabrication process. With advanced fabrication techniques being developed, researchers have already observed some shifts in the magic angle range of twisted BLG [109] by fabricating more refined devices. There's an opportunity to enhance the fabrication methods in this field, wherein utmost care should be taken to avoid any misleading results.

Nano-fabrication and growth of 2D materials often exhibit a range of defects such as single vacancy, Stone-Wales, nanopores etc. These effects can significantly influence the effective properties of the twisted nanostructures. Moreover, the accuracy and quality of the samples are often dependent on the type of synthesis method along with different other calibration and control parameters during the process. Detailed investigations are required in these directions to quantify the influence of manufacturing defects and other uncertainties concerning the process parameters.

The scalability of bilayer twisted graphene and other nano-heterostructures is a challenge, as it is difficult to fabricate these materials on a large scale while maintaining their desired properties and geometric configurations. Such crucial aspects need immediate attention for industrial-scale adoption of the knowledge generated in research laboratories.

7.9. Service-life effects: environmental and operational conditions

Besides manufacturing uncertainties, the service-life conditions of twisted 2D materials and other heterostructures need significant attention. Effects of the surrounding environment, material degradation, and accumulation of damage over time should be included in the computational design process in line with the notion of digital twins. Moreover, there are a few critical long-term mechanical properties such as fatigue, creep, and the effect of synthesis methods in the failure modes, which have not received adequate attention yet. Further research is required in such directions for a reliable adoption of these complex nanostructures in industry-scale applications.

8. Concluding remarks

Post the remarkable findings of unconventional superconductivity in twisted bi-layer graphene, wide attention is drawn to 2D van der Waals twisted multi-layered heterostructures. The coupled design space of stacking and rotating of layers, gives the hybrid and engineered superlattices entirely different properties than those of 2D monolayer structures. The rotation of a layer with respect to another layer in a 2D van der Waals heterostructure leads to various periodic moiré superlattices and combining it with the interlayer couplings between the layers results in exotic electronic, optical, magnetic and mechanical properties. In this article, we have started with the formation of electronic band structure in twisted BLG exhibiting flat bands around the 'magic angle'. The findings of superconductivity and other exotic properties in twisted graphene layers have opened a new way of looking at correlated physics in a range of other twisted 2D material configurations, the progress of which are subsequently discussed here. Along with twists, heterostrain is shown to largely

influence the electronic band structure and the flat band regime, and so eventually the optical and electronic properties.

Enlightened by the uniqueness in the physics and unprecedented properties of magic angle twisted BLG, there exists an extensive opportunity to explore multi-layer graphene and other 2D heterostructures with interlayer twisting to enhance and tune the electronic properties along with other simultaneous multi-objective goals. We have noted that it will be compelling to explore the addition of other prospective design parameters such as layer-wise stretching and heterostrain, interlayer shifting and translation, pressing, different forms of programmed defects and a combination of them with interlayer twisting on the tunable multiphysical properties of engineered 2D materials. The prospective role of artificial intelligence and machine learning therein is discussed for exploring the vast scope of combining different variants of 2D materials with numerous possibilities of stacking sequences along with the aforementioned expanded design space for identifying target combinations of the parameters that would have been impossible to explore solely using conventional methods of simulations and experiments. In this context, the aspect of physical realization of computationally invented configurations is duly emphasized. Due to the engineering at a nano-length scale and being atomically thin, it is quite complicated to fabricate single and multi-layer 2D materials and their heterostructures. Moreover, providing the accurate amount of layer-wise twists along with other proposed design-level parameters like programmed defects, direction-dependent strain, and translation would demand a high level of precision in the fabrication process.

In summary, through a concise review of the existing literature and current trends, we highlight that the field of twisted graphene and heterostructures is poised for significant growth and development in the foreseeable future, and there are numerous exciting opportunities to explore the rich and coupled design space for achieving unprecedented physical properties including a multi-objective target output space with application-specific assignment of weights by infusing the notion of engineering judgment.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

KS would like to acknowledge the doctoral scholarship received from Ministry of Education (MoE), India. T M acknowledges the initiation grant received from the University of Southampton.

ORCID iDs

S Naskar https://orcid.org/0000-0003-3294-8333
T Mukhopadhyay https://orcid.org/0000-0002-0778-6515

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