

Electron quantum metamaterials in van der Waals heterostructures

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In recent decades, scientists have developed the means to engineer synthetic periodic arrays with feature sizes below the wavelength of light. When such features are appropriately structured, electromagnetic radiation can be manipulated in unusual ways, resulting in optical metamaterials whose function is directly controlled through nanoscale structure. Nature, too, has adopted such techniques—for example in the unique colouring of butterfly wings—to manipulate photons as they propagate through nanoscale periodic assemblies. In this Perspective, we highlight the intriguing potential of designer structuring of electronic matter at scales at and below the electron wavelength, which affords a new range of synthetic quantum metamaterials with unconventional responses. Driven by experimental developments in stacking atomically layered heterostructures—such as mechanical pick-up/transfer assembly—atomic-scale registrations and structures can be readily tuned over distances smaller than characteristic electronic length scales (such as the electron wavelength, screening length and electron mean free path). Yet electronic metamaterials promise far richer categories of behaviour than those found in conventional optical metamaterial technologies. This is because, unlike photons, which scarcely interact with each other, electrons in subwavelength-structured metamaterials are charged and strongly interact. As a result, an enormous variety of emergent phenomena can be expected and radically new classes of interacting quantum metamaterials designed.

Electrons in solids have many different faces. For example, the diffraction of the electron wavefunction as it propagates through a material creates a complex pattern that determines its electronic properties (Fig. 1a). At the same time, an electron is a charged particle and possesses an electric field distribution that enables it to interact with other particles (Fig. 1b); this underlies its correlated behaviour. In naturally occurring materials, these various wavefunction and Coulomb characteristics are fixed by the particular crystal structure and material environment. As a result, the *modus operandi* for the discovery of new behaviour in electronic systems often requires surmounting the challenges of growth and synthesis of completely new materials and systems.

An alternative, and rapidly progressing, nanotechnology is that of van der Waals (vdW) heterostructures¹, wherein stacks of atomically thin materials are assembled to form a complex quantum system. This has been driven from multiple angles, including the proliferation of available atomic-layered materials^{1–3} over a wide range of phases such as semiconductors, semimetals, superconductors, ferromagnets and topological insulators; means of assembling high-quality vdW stacks (for example through mechanical pick-up/transfer assembly^{4,5}); and theoretical proposals of how these materials, when combined, can conspire to produce new physical phenomena. These two-dimensional (2D) materials are a particularly attractive putty to use in moulding materials-by-design because their electronic states are fully exposed and thus easily addressable. Few, if any, limitations are placed on the constituent layers, as each layer is relatively stable in-plane¹. Moreover, stacking such layers allows a far larger range of combinations than is conventionally possible by traditional epitaxial methods. All of these factors have led to a dizzying growth in the complexity of atomic-layered stacks and diverse research avenues⁶.

How should we view the progress in vdW heterostructures¹ to better navigate the path ahead? Here, we frame this rapid progress in terms of creating quantum metamaterials for electrons. As we discuss below, atomic-layered stacks provide a powerful means of

directly engineering the separate facets or character of electrons in solids, such as the texture of quasiparticle wavefunctions or the Coulomb fields that a charged particle possesses (Fig. 1). This capability arises from the very small feature sizes (such as lateral feature size a and vertical distance d ; see Fig. 1c) that are naturally formed in atomic-layer stacks. Crucially, these features are tunable over multiple length scales that range from distances much smaller than, and up to, the characteristic length scale that defines each of the electron's aspects—for example, the electron wavelength as well as the unit cell characterizes a quasiparticle's quantum wavefunction; the screening length defines the scale over which Coulomb fields vary. Tuning these lateral and vertical features enables strong modification of electron behaviour, in loose analogy to optical metamaterials; the electron quantum metamaterials that we discuss below use both wavelength- (or characteristic length scale) and subwavelength-scaled vdW structures. This is vividly illustrated in a prototypical stack of van der Waals materials (Fig. 1c) in which a simple twist of the layer orientation allows nearly continuous variation of features on scales smaller than the electron wavelength (for example atomic-scale registration toggled by twist angle) as well as wavelength-scale features such as the size of the superlattice unit cell.

In the following, we lay out how vdW structure in electron quantum metamaterials with feature sizes at or below the various characteristic (electronic) length scales in vdW stacks can be used as design strategies for engineering quantum behaviour. As we will see below, these strategies collectively fall into a length-scale engineering toolbox for emerging nanotechnologies—an intuitive framework for fundamental and technological progress.

Atomic-layer twists and turns

In mathematics, textiles and art, intricate spatial interference configurations—the familiar moiré patterns—can be formed when two periodic templates are overlaid. Similarly, when two atomic 2D layers are stacked on top of each other, the periodic atomic structure within the constituent layers combines to create new spatial

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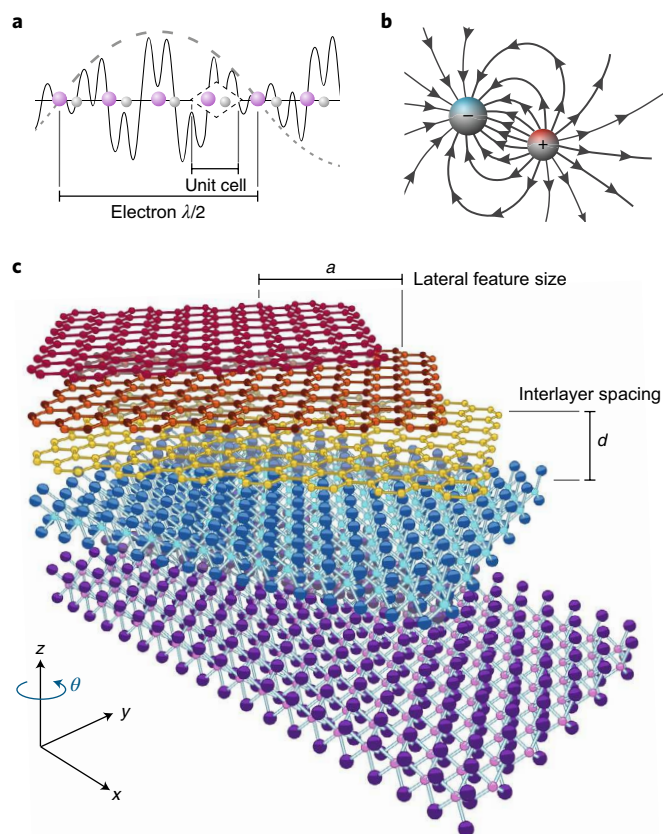


Fig. 1 | Wavefunction and Coulomb characteristics of electrons in vdW heterostructure quantum metamaterials. **a**, Schematic of the electronic wavefunction. The basic building block of the crystal, or crystal unit cell (outlined by dashed blue line), introduces repeating structure that modifies the electron matter wave of wavelength λ . **b**, Schematic of the electric field lines, a visualization of the local Coulomb field, surrounding an electron in proximity to a positive charge. **c**, A vdW heterostructure quantum metamaterial composed of individual 2D layers (TMDs, graphene and BN) and characterized by lateral feature size a , interlayer spacing d and atomic-layer twist angle θ .

patterns (Fig. 2) that resemble artificial lateral superlattices. These naturally possess sub-electron-wavelength features and offer a means of directly engineering the crystal unit cell (Fig. 2). When two 2D layers are commensurate^{7–12}, the electrons can be described exactly by new Bloch bands with an expanded unit cell defined by the superlattice structure (Fig. 2a). Even when the two layers have an incommensurate configuration, their moiré pattern¹³ allows the low-energy quasiparticle excitations to be effectively described by a set of moiré-superlattice Bloch minibands^{7–11,14–16}. Here we use the same language of effective Bloch bands and effective unit cells to describe both cases.

Engineering unit cell size. Together with control over the unit cell, in particular its size, superlattices also possess an associated (reduced) Brillouin zone^{7–11,14–16} that is tunable. The resultant Bloch minibands can be understood as bands folded in at the edges of the superlattice Brillouin zone. This vastly enriches the original band structure and grants control over the pattern of electron and hole filling^{15,17–20} (the free carrier density and type) in the heterostructure. A striking example is the set of secondary Dirac cones formed from the moiré pattern in graphene/hexagonal boron nitride stacks^{15,17–20} (G/hBN). In addition to a primary Dirac cone, replica Dirac particles are displaced at a higher energy defined by the

superlattice and exhibit tunable ambipolar behaviour as the Fermi energy passes through the secondary charge-neutrality points^{17–20}. Twisted bilayer graphene provides a particularly arresting illustration in which a rich array of minibands form and morph as a function of twist angle^{7,9–11,21}.

The facility to engineer unit cell sizes and its electron filling can lead to remarkable results when a magnetic field is applied: for example, a fractal Landau-level spectrum (Hofstadter butterfly) can be observed^{17–19}. The moiré superlattice sizes accessible in atomic-layer heterostructures make such intricate behaviour readily observable at realizable magnetic field strengths. This interplay between magnetic flux and superlattice size can even persist at high temperature: for example, Brown–Zak magnetic oscillations²² (Fig. 2a).

Texturing the unit cell. In optical metamaterials, sub-wavelength-scale features alter the propagation of light. In electronic materials, structure that arises within the unit cell (the relative amplitudes and phases of the electron wavefunction as it passes through each of the atoms or orbitals of the crystal) provides a patterned texture to the periodic part of the Bloch wavefunction. This texture is responsible for quantum geometric (wavefunction) properties such as non-trivial pseudo-spinor texture, Berry curvature and electric polarization²³, and can mediate nonlinear responses^{24,25}. The expanded unit cells innate to layered heterostructures (Fig. 2b) provide a natural playground in which to design Bloch wavefunctions from the bottom up. In these, the specific atomic configuration can be tuned by twist angle, relative lattice constants or the composition of each layer, as well as the field effect.

Bilayer stacks with unit cells comprising atoms from both top and bottom layers are prime targets for texturing. A well-known example is (Bernal-stacked) bilayer graphene, in which the linearly dispersing Dirac dispersion in each of the constituent layers hybridizes to form a spectrum of massive charge carriers. Similarly, in trilayer stacks, the band structure morphs, possessing bands that can intimately depend on its stacking arrangement²⁶ (for example ABA versus ABC). In much the same way, the heterobilayer G/hBN develops bandgaps near the Dirac point¹⁹ owing to AB sublattice asymmetry in the superlattice unit cell.

Perhaps the most striking consequence of atomic-layer twisting is that it enables us to produce new pseudo-spinor textures that are radically different from those found in the individual 2D layers. Whereas the pseudo-spin of an electron wavefunction in graphene simply winds around the equator in a Bloch sphere, the pseudo-spin vector in G/hBN or biased bilayer graphene cants out-of-plane (Fig. 2b) and permits a non-zero Berry curvature to develop close to band edges²⁷; Berry curvature encodes an intrinsic orbital angular momentum of wavepackets in the band structure induced by the configuration of atoms and orbitals in the unit cell²³.

The patterned wavefunctions of twisted vdW heterostructures can provide a unique venue in which to control Berry phase effects that result from pseudo-spin texture²³. Accessing the unusual quantum transport enabled by non-trivial wavefunction texture (quantum geometry) is currently being explored in many material systems such as Weyl semimetals. Progress, however, is no more evident than in vdW heterostructures, where Berry curvature can mediate a wide variety of unconventional quantum geometric effects that include valley-selective Hall transport^{27,28}, non-local resistance^{20,29,30} and valley (orbital) magnetic moments^{27,31}, to name a few. Quantum geometric effects can be particularly sensitive to the underlying symmetry of the crystal²⁴ wherein lowered crystal symmetry (engineered by strain, for example) can lead to unusual effects such as a current-induced (orbital) magnetization³², or a non-linear Hall current²⁴ even at zero applied magnetic field.

The expanded unit cells in vdW stacks are also characterized by additional degrees of freedom (DOF). When top and bottom layers are symmetric, as in bilayer graphene, polarization of each layer acts as an additional DOF. Breaking this symmetry, either by an external

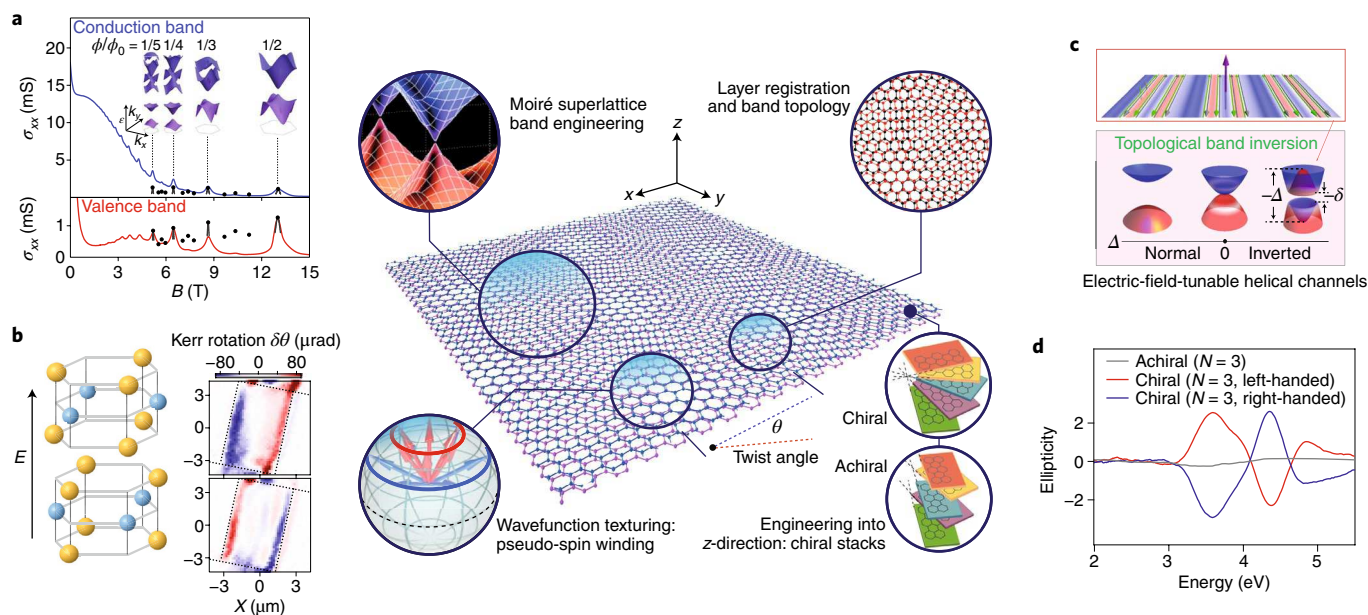


Fig. 2 | Wavefunction engineering in vdW heterostructures. **a**, Expanded unit cells in moiré superlattices yield a new pattern of Bloch minibands (circular inset). Superlattice unit cell size and magnetic flux can conspire to produce Brown-Zak magnetic oscillations (peaks shown in the plot) in G/hBN superlattice that can persist to high temperature. **b**, The texture, comprising the relative phases and amplitudes of the electron wavefunction on the orbitals or atoms in the stacked unit cell, can take on a non-trivial pseudo-spin winding (circular inset). Bloch-band Berry curvature can be unlocked in bilayer MoS_2 by applying an out-of-plane electric field that breaks inversion symmetry. This manifests as a valley Hall effect, with valley density accumulating at sample edges producing a Kerr rotation. **c**, The local stacking configuration can influence the Bloch-band topology (circular inset). When the local stacking registration changes spatially, 1D (topological) kink states can manifest; in a TMD heterobilayer, these helical kink states can be switched on or off by an electric field. **d**, Stacking arrangement of vdW layers (for example handedness, or chiral versus achiral) can program ellipticity (circular dichroism) directly into the stack. Adapted from ref. ²², AAAS (**a**); ref. ³¹, SNL (**b**); ref. ⁴⁶, SNL (**c**); ref. ⁵⁷, SNL (**d**).

electric control^{33,34} or by spontaneous means^{35,36}, induces new phases³⁷. The unit cell also embodies the symmetries of the larger heterostructure. For example, whereas monolayer MoS_2 breaks inversion symmetry, bilayer MoS_2 preserves it; an additional out-of-plane electric field is required to break inversion symmetry³¹ and unlock the Berry curvature of the bands (Fig. 2b). This interplay of symmetry and the expanded DOFs becomes particularly striking when spin and valley (tied to magnetization) and layer (tied to electric polarization) DOFs are strongly coupled³⁸, enabling magneto-electric coupling and electrical access to inner DOFs.

Topological bands. Our ability to texture the Bloch wavefunctions in 2D twisted quantum metamaterials informs a unique possibility: designing topological bands out of trivial materials. Indeed, many 2D materials (such as graphene, MoS_2 , hBN or bilayer graphene) possess a Dirac-type electronic structure closely related to that found in topological insulators³⁹. This ‘topological transmutation’ can take one of several forms (Fig. 2c). For example, the stacking arrangement between top and bottom layers can slip at stacking faults. This sudden change in the crystal structure yields contrasting unit cell configurations across the fault line. In gapped bilayer graphene, such stacking faults host localized topological domain-wall states, which are gapless and valley-helical^{40–42}; domain-wall kink states can also be achieved by electric field using split-dual gates^{43,44}. In 2D heterobilayers, manipulating the unit cell texture can enable the design of topological bands in commensurate G/hBN⁴⁵ and TMD bilayers⁴⁶. Remarkably, when the atomic registration is incommensurate, 1D (helical) kink states can proliferate and form a network-like structure that can be switched on or off by an electric field⁴⁶ (Fig. 2c) and reconfigured spatially by strain.

Switchable topological bands, for which we can electrically turn the helical edge states on or off, also extends to intrinsic

atomic-layer topological insulators⁴⁷, such as that found recently^{48–50} in $1\text{T}'\text{-WTe}_2$. The layered nature of vdW topological insulators means that they can be easily stacked, providing a straightforward means of parametrically increasing the edge conductance⁴⁷; stacking can also make it possible to construct helical modes by combining edge states of opposite chirality in an electron–hole bilayer in the presence of a magnetic field⁵¹.

Excited states and quantum colouring. Although so far we have considered only the lowest-energy excitations, the higher-energy collective modes of quantum metamaterials can also be tuned by sub-electron-wavelength features. Take, for example, an exciton, which is normally considered a simple (hydrogenic-like) bound state of an electron and hole. In conventional materials, the narrowly defined exciton energy gives rise to sharply resonant optical emission and absorption. In the presence of a unit cell structure with features smaller than the Bohr radius, the constituent electron and hole may possess Berry curvature. Because of this, the exciton optical spectrum morphs, exhibiting split angular momentum states^{52,53}. The resultant change in the optical absorption is a unique quantum effect resulting from sub-electron-wavelength features.

Similarly, plasmons—the collective modes formed from a high density of charge carriers—may experience chirality at zero-magnetic field^{54,55} if the constituent electrons or holes possess non-trivial Berry curvature. These are some examples of how collective modes may take on beyond Landau-Fermi liquid type characteristics⁵⁶ induced by non-trivial unit cells.

Length-scale engineering into the third dimension. Beyond two layers, further stacking in the out-of-plane direction (three, four and more layers) provides even more opportunity to engineer the structure and wavefunction texture of the unit cell. One interesting

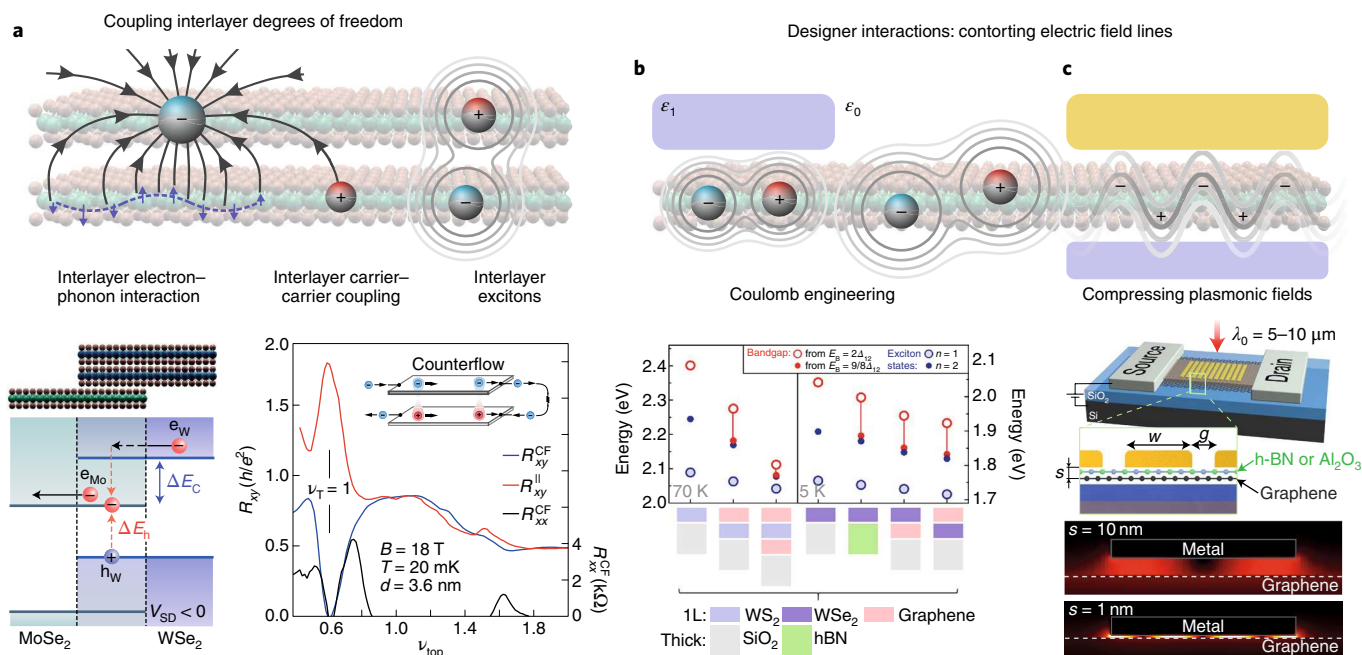


Fig. 3 | Designing interactions in vertical vdW stacks. **a**, The z -extent of Coulomb fields enables charge carriers in one layer to couple or interact with other DOFs in an adjacent layer (top). These DOFs include phonons, other charge carriers and excitons. Efficient interlayer electron-hole multiplication is generated from strong interlayer carrier coupling (bottom left). Electrons and holes in separated vdW metallic layers can condense to form an excitonic fluid characterized by a quantized Hall drag^{69,70} (not shown), as well as a dissipationless nature (see results for counterflow geometry close to total filling $\nu_T=1$) in a double-bilayer graphene heterostructure (bottom right). CF, counterflow. **b**, Excitons possess Coulomb field lines that extend out of the 2D plane and are sensitive to environment adjacent to the 2D material (top). These enable exciton spectra as well as the bandgap to be modified by the dielectric environment (bottom). Heterostructure comprises single layer vdW materials ("1L") stacked with a dielectric substrate ("Thick"). **c**, Similarly, the z -extent of a plasmon's electric field (as well as its in-plane charge distribution) is sensitive to the surrounding environment, allowing a nearby metallic plate (yellow) to slow the plasmon down^{74,75} (top), or to compress the z -extent of its electric field⁷⁶ (bottom) creating electromagnetic mode volumes up to 10^9 times smaller than that found in free-space. Adapted from ref. ⁶³, SNL (**a**, bottom left); ref. ⁷⁰, SNL (**a**, bottom right); ref. ⁷², SNL (**b**); ref. ⁷⁶, AAAS (**c**).

prospect is how motion in the out-of-plane direction can be coupled to motion in the in-plane directions. Inspiration may come from gyrotropic optical media, in which left- and right-elliptical polarizations can propagate at different speeds through the material. Such handedness can, for example, be used to determine the chirality of molecules by measuring the change in linearly polarized light upon transmission. Carefully stacking and combining small atomic twists in a vdW heterostructure could realize such optical handedness in a 3D crystal structure⁵⁷ (Fig. 2d). There are proposals for chiral materials composed of self-assembled inorganic materials⁵⁸, but the electronic or optoelectronic behaviour of such assemblies remains under investigation⁵⁹.

Designer interactions in the solid state

Unlike photons, which travel independently through an optical medium, electrons may interact strongly in low-dimensional materials. While physically confined to move within the 2D x - y plane, electrons in vdW materials are charged particles that possess electric fields extending out in all directions: in particular, the out-of-plane or z -direction. This is a direct result of the 3D nature of the Coulomb interaction. Indeed, the extended z -direction profile of an electron's potential enables the electric field effect⁶⁰—a ubiquitous means of controlling the carrier density in a 2D material by a proximal gate electrode. This Coulomb-field character of the electron (Fig. 1b) is responsible for a rich tapestry of phenomena, from scattering with phonons and impurities to bound electron-hole pairs and exotic correlated phases.

For vdW materials, vertical stacking (Fig. 1c and Fig. 3) enables the engineering of features that are far smaller than the characteristic length scales governing the electron's Coulomb fields.

The exposed surfaces (and surface electronic states) of vdW materials make them particularly susceptible to the environment. Further, separate vdW layers can be stacked flush against each other or, when electrical isolation of the layers is required, thin hBN spacer layers (as narrow as several atomic layers) can be used. This is in stark contrast to conventional bulk materials, in which electrons residing deep in the bulk dominate the material properties. As a result, the in-plane motion and dynamics of charges can be manipulated by their Coulomb fields out-of-plane.

'Stray' fields and layer-to-layer interactions. Coupling between in-plane DOFs with out-of-plane DOFs are an immediate way that these 'stray' Coulomb fields affect electronic behaviour. A case in point is the coupling of in-plane electrons with phonons in a nearby substrate or dielectric medium (Fig. 3a). Phonons in the surrounding medium, characterized by the displacement of lattice ions, possess dipoles that respond readily to extended electric fields of the electron. While this coupling is present in all heterostructure stacks, the fully exposed electric potential profile of 2D layers allows it to be taken to the extreme: for example, in insulator/graphene/insulator stacks, the scattering of electrons by phonons can be dominated by the surrounding dielectric. Indeed, G/hBN stacks have been shown to enable fast cooling of electrons in graphene by hyperbolic phonons in hexagonal boron nitride^{61,62}.

Stray-field coupling extends far beyond electron-phonon scattering. The out-of-plane extent of electric fields can mediate interaction between a wide range of DOFs and quasiparticles even when they are in separate planes (such as excitons, phonon-polaritons and plasmons). For example, electrons propagating in one 2D layer may efficiently generate additional interlayer electron-hole

pairs⁶³ (Fig. 3a, bottom left) rather than emit phonons. This multiplication process probably results from the increased interaction cross-section between electron Coulomb fields and the reduced phonon density of states in 2D materials.

Interestingly, interlayer excitons that form in vdW material stacks exhibit a dipole moment that may be slightly canted away from the z -direction owing to interlayer twisting^{64,65}. The relaxation (or excitation) of such canted interlayer excitons involves a well-defined phonon mode. Such a narrow phase space for exciton–phonon interactions could be used to engineer unusual devices that mimic vibronic transitions observed in photosynthetic energy transfer⁶⁶. Coupling of this sort can even grant unconventional transduction between other DOFs; for example, that of out-of-plane phonon-polaritons (charge-neutral objects), which can be tuned electrically by a proximal metallic layer when they hybridize with plasmon-polaritons⁶⁷.

Perhaps even more striking is how charge DOFs residing in two nearby metallic layers separated by a 2D insulator interact through their Coulomb fields, even when there is no particle exchange between them. For instance, inducing a current in one 2D layer can drag carriers in the adjacent layer by exchange of momentum and energy⁶⁸. When layer separation is smaller than the screening length, the strongly coupled regime (where interlayer interaction is as strong as intralayer interaction) can be accessed. For example, in this regime electrons and holes in the separated metallic layers condense to form an excitonic condensate^{69,70}—a phase of matter distinct from that of its constituent parts (Fig. 3a, bottom right). This is prototypical of an interacting quantum metamaterial and shows how the strong coupling of DOFs can lead to radically new behaviour.

How can the Coulomb field be engineered? Because field lines warp and contort depending on a vdW material's environment, stacking can provide the means to tailor the interaction between electrons. As an illustration, consider the simple structure of an atomically thin semiconductor (for example MoS₂) sandwiched on either side by insulators (Fig. 3b): the dipolar fields between an electron and a hole in the same layer get squeezed because of the strong contrast of dielectric constant between the monolayer and its surroundings⁷¹. This warping of field lines leads to a non-hydrogenic series of excited states for 2D excitons and a considerable modification of its bandgap energy⁷² which can be spatially engineered in such thin semiconductors (Fig. 3b, bottom).

Another simple structure consists of an atomically thin metal (such as graphene) placed very close to a bulk metallic plate (spaced by a 2D insulator). In such structures, the proximal bulk metal helps to screen electric fields caused by charge-density fluctuations in the atomically thin metal. When positioned closer than a screening length apart, the effect can be large. For example, a proximal graphite gate can smoothen out charge puddles in graphene, leading to a more homogeneous carrier doping landscape^{19,73}. Using similar approaches, plasmons in graphene/insulator/metal heterojunctions can be slowed down^{74,75}, and plasmon electric field profiles in the z -direction can be squeezed into the plane⁷⁶ (Fig. 3c).

The warping and screening of Coulomb field lines can be taken to the ultimate limit: transforming the nature of the Coulomb interaction itself. One of the most remarkable proposals is to transmute the Coulomb repulsion (in a low-dimensional system) into an effective Coulomb attraction, by placing it close to a highly polarizable medium⁷⁷. In the early proposal of this⁷⁷, electrons in the polarizable medium form a 'glue' that mediates attraction in a low-dimensional conducting system; this can lead to a synthetic non-phonon-mediated superconductivity. Strikingly, very recent evidence of synthetic electron attraction from Coulomb repulsion has been observed⁷⁸. Because 2D conductors can be fully embedded in vertical stacks, yet have their field profiles fully exposed, vdW heterostructures can provide a unique platform to realize synthetic superconductivity^{79,80}.

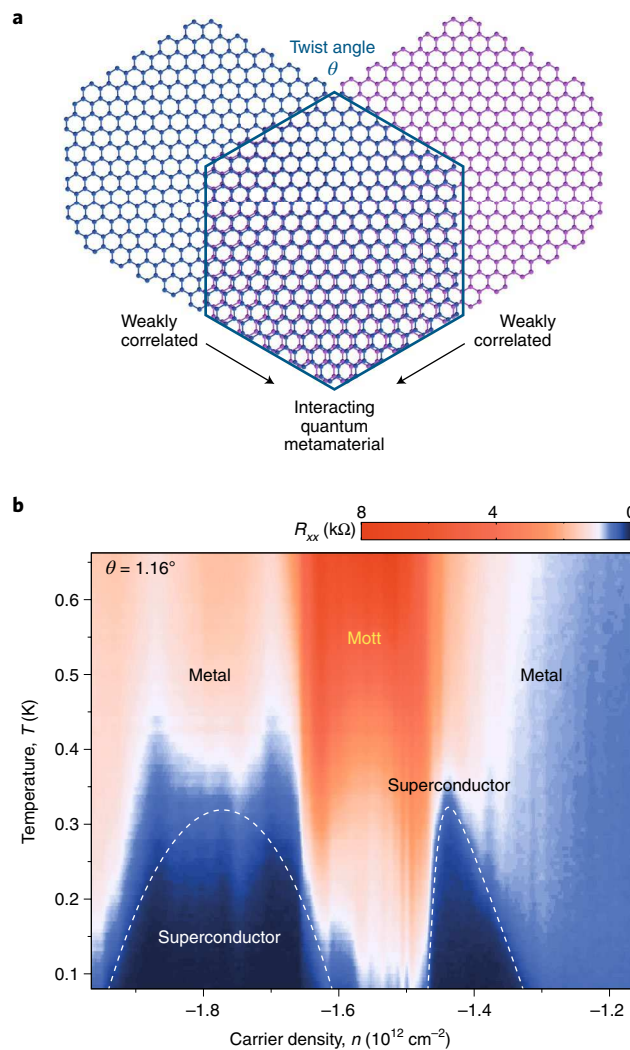


Fig. 4 | Strong correlations in interacting quantum metamaterials. **a**, The lateral structure formed in vdW heterostructures can work synergistically with electron interactions to produce emergent correlated phases. Single-layer graphene by itself (blue lattice or magenta lattice) is weakly correlated, but when they are combined (overlapping region), this gives rise to an interacting quantum metamaterial (conceptual illustration). **b**, Strongly correlated phases^{107,108} have been found in twisted bilayer graphene when the twist angle θ between layers is close to certain magic angles. These phases can be readily tuned by means of a gate, from a correlated insulator (here labelled 'Mott') to a superconducting phase and then to a metallic phase. Adapted from ref. 109, SNL (**b**).

The tools of the trade in quantum metamaterials. The designer-interactions toolkit described here outlines some emerging methods to control electron wavefunctions or the Coulomb fields of charge carriers. These tools principally exploit features below and up to an electron's characteristic length scales (Fig. 1a,b) to engineer artificial quantum material behaviour. Such tools, we believe, can be applied to other characteristic length scales and a wide range of as-yet-unexplored nanotechnologies. Given the myriad of correlated phases recently uncovered in 2D materials (for example, gate-tunable superconductivity in monolayer MoS₂ (ref. 81), ferromagnetism in monolayer Cr₂Ge₂Te₆ (ref. 82) and CrI₃ (ref. 83), and anti-ferromagnetism in CrI₃ stacks⁸³) the designer-interactions toolkit also provides new opportunities for tailoring and controlling correlated phases, particularly when distinct phases are proximal to each other^{84–86}. Electric-field control is one of the most compelling

Table 1 | Summary of key quantum metamaterial strategies for van der Waals heterostructures discussed in this Perspective

vdW heterostructure tools	Key concepts/strategies	Electronic properties	Examples of accessible phenomena
Lateral/vertical structure Atomic-scale registration controlled, for example, by twist, stacking alignment, choice of homo/hetero-bilayers, strain	Wavefunction texturing	Quantum geometry and Berry phase	Valley Hall effect
		Unit cell symmetry	Magneto-electric effect
		Excited-state internal structure	Chiral Berry plasmons
		Topological bands	Topological bands from trivial materials
		Internal magnetic degrees of freedom	Proximity induced spin-orbit coupling
		Chirality	Stacking-tuned dichroism/tunnelling
Vertical structure Proximal gate	Unit cell size engineering	Bandwidth	Magic-angle nearly flat bands
		Layer of degree of freedom	Electrically controlled gap
		Superlattice Bloch minibands	Secondary Dirac cones
Proximal dielectric stacks	Gate/field effect	Electron filling and density	Hofstadter butterfly
		Electron-phonon interaction	hBN-hyperbolic phonon cooling
	Stray fields and interlayer coupling	Carrier-carrier scattering	Coulomb drag
		Field extent and profile	Exciton/plasmon engineering
	Coulomb field engineering	Interaction sign	Synthetic superconductivity
Example of a combination strategy			
Wavefunction texturing + Gate/field effect		(Low) unit cell symmetry, spin degree of freedom	Electrical control over spin texture and Berry curvature in a monolayer TMD

attributes of using vdW correlated phases, as it can enable unusual couplings to arise: for example, bilayer CrI₃ exhibits an electric-field switchable magnetic order^{87,88}.

Our design list is not exhaustive: electrons have numerous other characteristics that can be directly engineered. A particularly conspicuous aspect is the spin DOF. This, too, can be directly tailored by proximity coupling to a strong spin-orbit coupling layer^{89,90} or a ferromagnet⁹¹. Another aspect is the vertical tunnelling characteristic of stacks that can be tuned by layer arrangement^{92,93} or magnetic ordering^{94–97}. Stacks may also exhibit structural phase transitions when the twist angle is small¹² or have layer alignment that is dynamically controlled⁹⁸; when coupled with electronic behaviour, these can lead to designer responses^{98,99}. Lastly, we remark that stacking different vdW materials on top of each other is by no means the only way to create features in vdW heterostructures. For example, lateral superlattices can be engineered by patterning a vdW material with a local top-gate, by stacking a vdW material atop a pre-patterned dielectric substrate^{100,101}, or by strain (see for example, ref. ¹⁰²). Patterned dielectric superlattices¹⁰¹ are particularly attractive because they enable high-quality lithographically defined superlattices possessing an electrically tunable miniband structure¹⁰¹ with the attendant properties of the superlattices discussed above; this also grants access to other types of structures (for example highly anisotropic 1D superlattices¹⁰⁰).

Our Perspective collects early examples in this rapidly developing research direction and distils from them emerging design strategies for electronic control in vdW heterostructures. Table 1 sketches how each of the vdW heterostructure (experimental) tools (column 1) enables control over electronic properties (column 3) through overarching key design principles (column 2). The design principles act as quantum metamaterials strategies for vdW

nanoengineers and provide a rational path for tailoring material properties. Given that many electronic properties (as well as strategies) are interlinked, one striking application of the design principles listed is to use the principles in tandem. Such combination strategies can enable new functionality (see Table 1): for example, low unit cell symmetry can conspire with the field effect to grant electrical control over the internal quantum structure of electrons, such as the spin structure (including out-of-plane component) and quantum geometry of transition metal dichalcogenides (TMDs; see refs ^{103–106} for a recent example in monolayer 1T'-WTe₂).

Interacting quantum metamaterials

One of the most promising aspects of quantum electronic metamaterials—and one that sets them wholly apart from their photonic counterparts—occurs when electrons in subwavelength-structured samples interact to exhibit unexpected emergent behaviour (Fig. 4). Such systems potentially provide a venue for a variety of complex emergent phenomena that arise from relatively simple constituents. A simple illustration of the effect of cooperation between subwavelength features and interactions can be found in G/hBN (twisted) moiré superlattices. At the single-particle level, the AB sublattice asymmetry (corresponding to an energy gap at the Dirac point) induced in graphene is expected to be minute, owing to the alternating gap sign in adjacent superlattice domains¹⁴. Yet because of the same superlattice structure, theory predicts that Coulomb interactions can renormalize the bare gap size with a faster scaling exponent (as compared with graphene without a superlattice structure), greatly enhancing the gap size¹⁰⁷.

Perhaps the most compelling landscape for interacting quantum metamaterials is that of magic-angle twisted bilayer graphene

heterostructures^{108,109}, in which a set of new correlated phases has been found (Fig. 4). Whereas strong coupling between top and bottom graphene layers at low twist angles produces a pattern of Bloch minibands, when the twist is close to ‘magic angles’ the electronic structure is pushed to an extreme limit^{11,110}: nearly flat electronic bands are formed. As a result, electron kinetic energy is quenched, and the interaction parameter increases. At half-filling, strong correlations between electrons localized near AA stacking regions yield an insulating gap consistent with that of a correlated insulator, such as a Mott insulator¹⁰⁸ (Fig. 4b). When doped away from half-filling, magic-angle twisted bilayer graphene becomes superconducting¹⁰⁹. This superconductivity exhibits doping-dependent domes reminiscent of high-temperature superconductors (Fig. 4b) and a critical temperature just under 10% of the Fermi temperature (indicative of strong coupling superconductivity).

What is the future of length-scale engineered nanotechnologies? The emergent behaviour in twisted bilayer graphene was a surprise; it is a remarkable demonstration of how electron interactions and subwavelength features conspire in interacting quantum metamaterials to produce radically new phases. Strikingly, this strongly correlated behaviour arose when two seemingly weakly correlated atomic-layered materials (graphene) were stacked together—a quantum alchemy of sorts. Similar correlated insulating behaviour has been observed in ABC trilayer graphene on hBN heterostructures¹¹¹. Together, these paint an exciting future for nanotechnology based on vdW heterostructures. What other transmutations are possible when atomic layers, each possessing already ordered (interacting) phases, are combined? The opportunities are vast.

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Competing interests

The authors declare no competing interests.

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