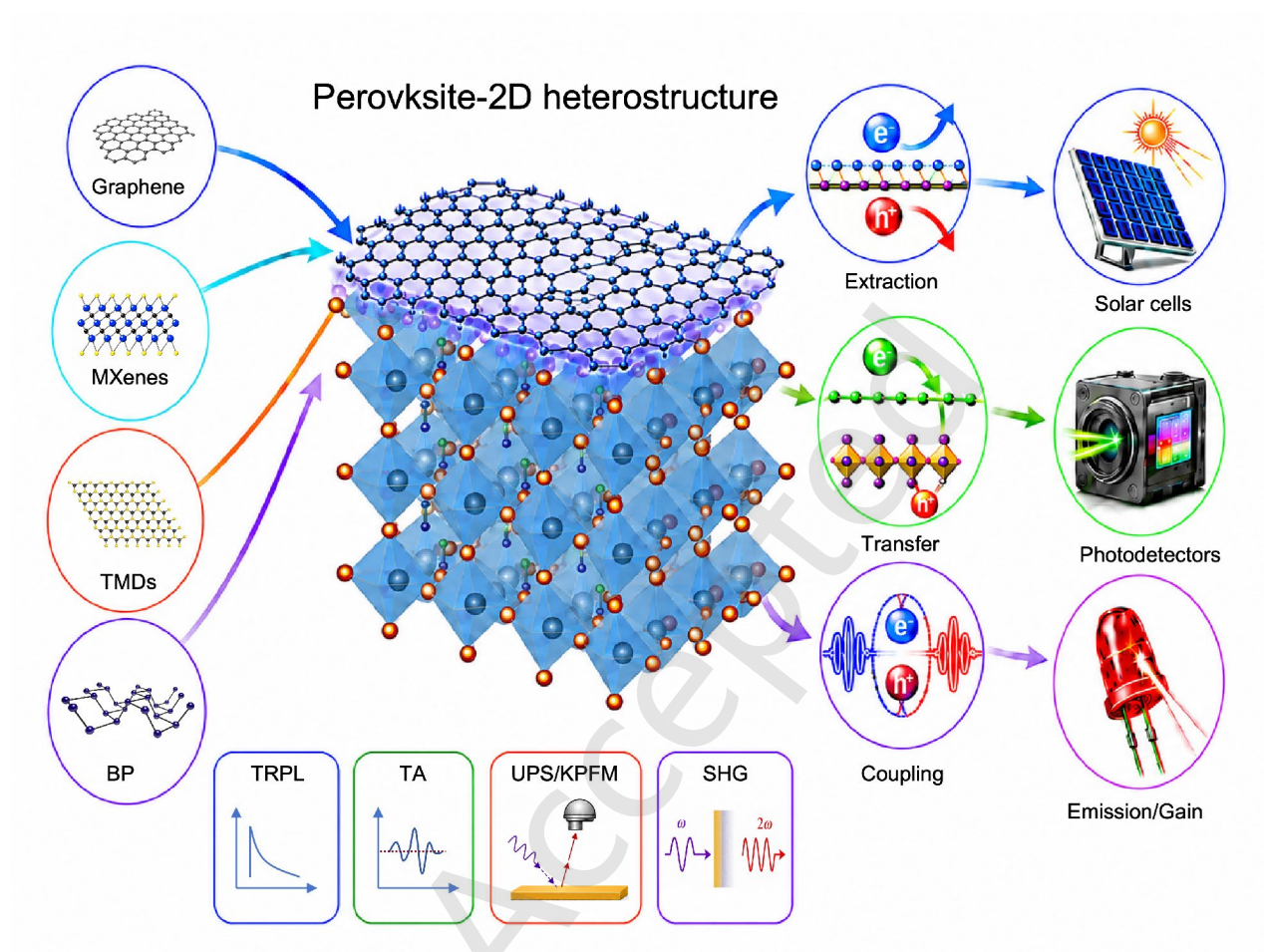


Graphic Abstract



This review centers on the interfacial photophysics governing the fate of photoexcited populations in halide perovskite–2D heterostructures. It distinguishes extraction-dominated, loss-dominated, and coupling-sensitive or radiative-state-preserving pathways, and highlights how TRPL, transient absorption, UPS/KPFM, and SHG provide complementary evidence for their mechanistic assignment. These photophysical pathways connect graphene-, MXene-, TMD-, and black-phosphorus-based interfaces to their consequences in solar cells, photodetectors, and emission or optical-gain devices.

Review

Interfacial photophysics in perovskite–2D materials heterostructures

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Abstract

Perovskite–2D material heterostructures have evolved from empirical interface engineering motifs into model systems for controlling interfacial photophysics in solution-processed optoelectronics. Graphene, MXenes, transition-metal dichalcogenides, black phosphorus and related layered materials can improve device operation through charge extraction, contact stabilization, recombination suppression, morphology control or radiative-state management. These outcomes, however, cannot be understood from material identity alone. This Review organizes perovskite–2D interfaces according to the interfacial variable they control and the photoexcited-state pathway they enable. Electrode-facing graphene and MXene contacts primarily regulate tunnelling, work-function alignment, carrier extraction and stability. Semiconducting perovskite/TMD heterostructures introduce finite-bandgap interfaces where selective transfer, interfacial coupling and excitonic or valley-sensitive responses become important. Structure- and field-mediating interfaces act through growth control, strain release, grain-boundary carrier pathways or local electrostatic fields rather than through uniform planar charge transfer. We then connect these archetypes to three interfacial photophysical regimes: extraction-dominated, loss-dominated and coupling-sensitive. Particular emphasis is placed on spectroscopic readout, including TRPL, transient absorption, UPS/KPFM, photocurrent dynamics and SHG or time-resolved SHG, because similar observables can arise from

different mechanisms. Finally, we discuss how these pathways determine solar-cell, photodetector and emission or gain-device performance, and outline future directions toward cleaner, more programmable and more diagnosable perovskite–2D interfaces.

Keywords: perovskite-2D heterostructures; interfacial photophysics; charge extraction; non-radiative recombination; interfacial coupling

1. Introduction

Halide perovskites have emerged as a versatile platform for both high-performance optoelectronics and the study of non-equilibrium excited-state processes in solution-processed semiconductors. Their strong optical absorption, long carrier diffusion lengths, defect tolerance, and broad compositional tunability have enabled rapid progress in photovoltaics, photodetection, and light emission. At the same time, it has become increasingly clear that the operation of perovskite devices is not determined by the absorber alone, but also by the interfaces through which photoexcited carriers are extracted, redistributed, trapped, or coupled to adjacent functional layers.^{1,2}

Within this context, two-dimensional materials were first introduced into perovskite devices largely from the perspective of interface engineering. Graphene, MXenes, transition-metal dichalcogenides (TMDs), black phosphorus (BP), and related layered materials have been employed as transparent contacts, conductive interlayers, selective transport components, or stabilizing interface modifiers, particularly in photovoltaic and photodetector architectures^{1–3}. This line of development has already demonstrated substantial technological value, including improved contact energetics, reduced interfacial loss, enhanced electrode robustness, and progress toward scalable module-level integration^{3,4}.

Equally important, however, is that perovskite/2D heterojunctions provide a setting in which the interface can be viewed as an active photophysical region rather than merely a passive transport boundary. Owing to atomic thickness, van der Waals assembly, tunable band alignment, modified dielectric screening, and reduced-dimensional interfacial geometry, the same interface may do far more than facilitate charge extraction. It may reshape recombination pathways, modulate local electrostatic and dielectric environments, alter interfacial symmetry, preserve or quench radiative excited states, and, in selected cases, support more coupling-sensitive excited-state behavior. In this sense, perovskite–2D heterostructures occupy a useful middle ground between classical interface engineering and more strongly coupled low-dimensional photophysics^{1,5}.

This broader perspective is what makes perovskite–2D interfaces particularly compelling as a research topic. A graphene-facing junction may still be understood primarily in terms of rapid extraction and electrical readout, whereas a semiconducting perovskite/TMD interface can raise qualitatively different questions involving selective transfer, interfacial coupling, or radiative-state-sensitive behavior. Likewise, some 2D-enabled interfaces may improve device performance mainly by reorganizing morphology, local fields, or interfacial disorder, rather than by introducing a simple

direct-transfer channel. The scientific challenge is therefore not only to report improved device metrics, but also to identify which interfacial photophysical pathway is actually operating and how it should be distinguished spectroscopically.^{1,2}

Previous reviews have extensively discussed perovskite interface engineering, 2D-material interlayers, and mixed-dimensional optoelectronic devices from the viewpoints of material selection, contact design, device architecture, and performance optimization. Here, we adopt a complementary photophysical perspective and focus on how perovskite–2D interfaces redistribute photoexcited populations among extraction, loss, and radiative or coupling-sensitive pathways. In this Review, we focus on heterointerfaces between halide perovskites and non-perovskite two-dimensional materials, with interfacial photophysics as the organizing principle. Examples drawn from related van der Waals or low-dimensional systems are provided as conceptual or methodological references to clarify transferable photophysical principles and spectroscopic criteria. Rather than attempting to catalogue all possible material combinations or interface phenomena, we explore how the 2D component changes the interfacial variable that controls the fate of photoexcited states. This perspective leads us to consider three dominant regimes: extraction-dominated interfaces, where photoexcited populations are converted into spatially separated or collectable carriers; loss-dominated interfaces, where they are redirected into trap-assisted or disorder-mediated non-radiative recombination; and coupling-sensitive interfaces, where radiative, excitonic, valley, or nonlinear optical responses are preserved or redistributed. Particular attention is therefore given to spectroscopic readout, because the central issue is often not simply whether an interface improves a device metric, but which interfacial excited-state pathway that interface brings into play. The discussion proceeds from interface archetypes to interfacial photophysical regimes and their spectroscopic signatures, and finally to their consequences for solar cells, photodetectors, and light-emitting or optical-gain devices, as summarized in Fig. 1.

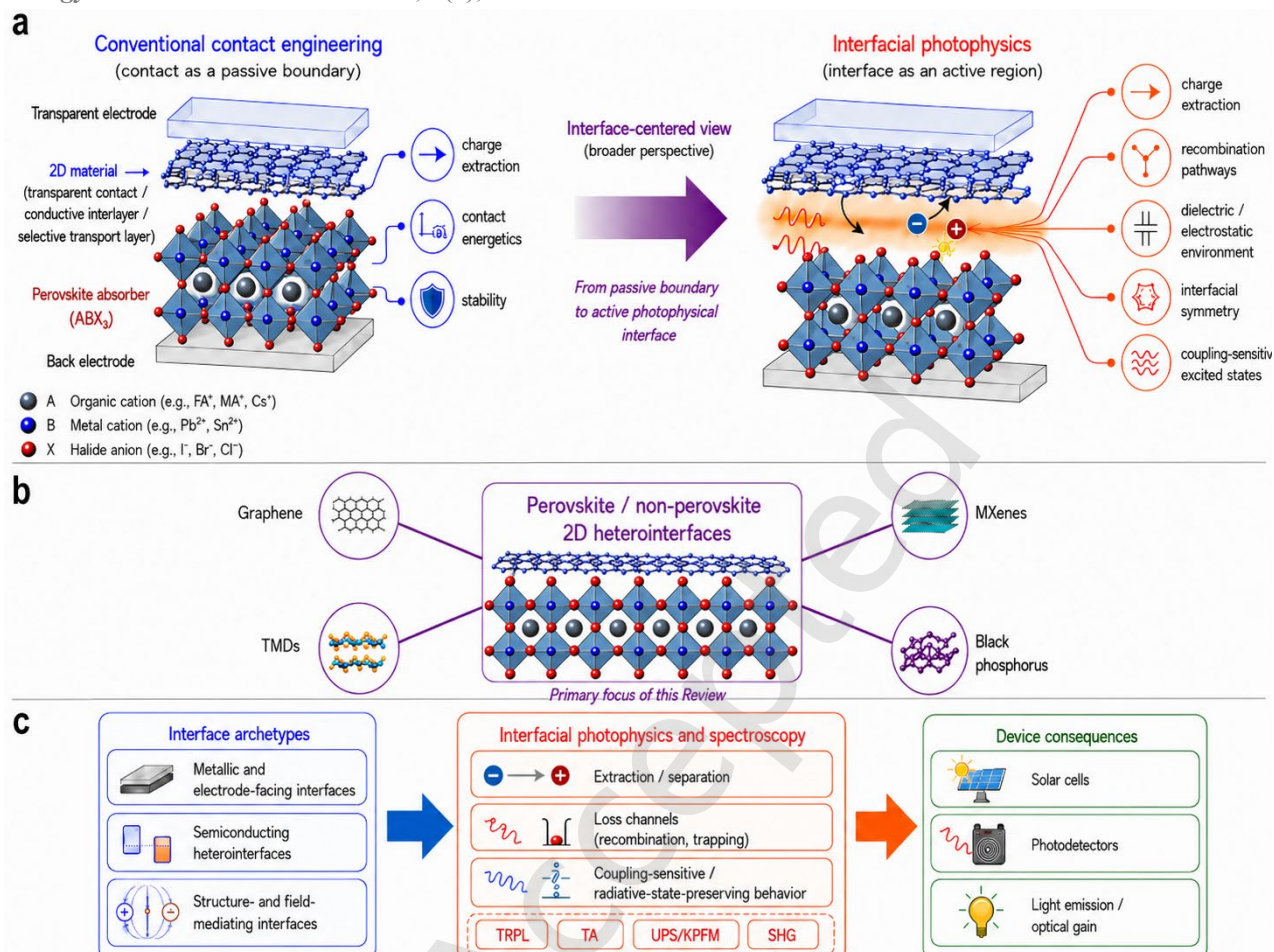


Figure 1 | From contact engineering to interfacial photophysics in perovskite–2D heterostructures.
 a, Conceptual shift from viewing the 2D layer primarily as a contact or transport component to treating the perovskite/2D junction as an active photophysical interface. b, Scope of this Review, focused on heterointerfaces between halide perovskites and non-perovskite two-dimensional materials, including graphene, MXenes, TMDs, and black phosphorus. c, Review logic linking interface archetypes to interfacial photophysical regimes and spectroscopy, and ultimately to device-level impacts in solar cells, photodetectors, and light-emitting or optical-gain devices.

2. Functional categorization of perovskite–2D heterostructures

Perovskite–2D interfaces are more usefully classified by the interfacial variable they control than by composition alone. As summarized in Fig. 2a–d, the representative archetypes discussed in this Review include electrode-facing graphene contacts, termination-controlled MXene interfaces, coupling-sensitive perovskite/TMD heterostructures, and buried field-mediated interfaces. Graphene and $\text{Ti}_3\text{C}_2\text{T}_x$ MXenes mainly define electrode-facing interfaces through contact continuity, tunnelling/extraction, electrode protection, and termination-dependent work-function tuning.^{4,6–8} TMD/perovskite heterostructures introduce finite-bandgap 2D crystals, enabling phase-controlled extraction, coupling-sensitive optical gain, and spin/valley-selective transfer.^{9–11} Black phosphorus adds an anisotropic finite-bandgap branch for direction-dependent transport and broadband response.^{12,13} Interfaces based on growth control, strain release, grain-boundary pathways, or local-field modulation form a separate structure- and field-mediating archetype, because their primary effect is not a uniform planar perovskite-to-2D charge-transfer junction.^{3,7,14,15} To clarify the function-based classification, Table 1 summarizes representative perovskite–2D interfaces across different material classes, interface geometries and device contexts. The selected examples are organized by their dominant interfacial photophysical role rather than by material identity alone.

Table 1 Representative perovskite–2D interfaces classified by interfacial photophysical function.

2D material class / representative systems	Representative interface geometry	Device context	Dominant photophysical role	Diagnostic evidence / endpoint	Representative studies
Graphene and graphene-derived contacts	Electrode-facing transparent contact; plane-contacted van der Waals junction; photogating channel	Solar cells; semitransparent or scalable panels; photodetectors	Extraction-dominated response or photogating-assisted electrical readout	Reduced injection barrier, gate-dependent tunnelling, ultrafast transfer signal, photocurrent or channel-conductance modulation	refs. 4, 6, 8, 15, 38
MXenes, including $\text{Ti}_3\text{C}_2\text{T}_x$ and Cl-terminated MXene interfaces	Perovskite/ETL contact; buried field-mediated interface	Solar cells and modules	Extraction-dominated or field-mediated separation	Work-function shift, band bending, interfacial dipole, improved charge collection, reduced recombination signatures	refs. 3, 17, 17, 18
TMDs, including MoS_2 , WS_2 and WSe_2	Semiconducting perovskite/TMD heterostructure; van der Waals epitaxial template; finite-bandgap 2D contact	Photodetectors; optical-gain and lasing devices; spin/valley optical response	Extraction-dominated transfer or coupling-sensitive excited-state response	Layer-specific TA response, phase-dependent extraction, optical gain or lasing threshold, valley polarization, excitonic or nonlinear optical signatures	refs. 9–11, 14, 18, 37, 39
Black phosphorus	Anisotropic finite-bandgap interface; local grain-boundary or transport pathway	Photodetectors; BP/perovskite detector interfaces; charge-collection interfaces	Extraction/loss competition governed by anisotropic transport and interfacial recombination	Photocurrent anisotropy, broadband photoresponse, transport-direction dependence, recombination or trap-related optical signatures	refs. 7, 12, 13, 16
Grain-boundary-localized 2D flakes, such as BP, MoS_2 and graphene oxide	Local flake-mediated pathway at perovskite grain-boundary edges rather than a continuous planar heterojunction	Solar cells and photodetectors	Loss modulation or local extraction depending on whether carriers are routed away from recombination-active boundaries	Spatial PL or TRPL contrast, local recombination changes, photocurrent response, trap-related or persistent-carrier signatures	ref. 7, 18, 19
BP quantum dots and other finite-gap 2D nanoflakes with endpoint evidence	Quantum-dot- or nanoflake-modified perovskite films or transport-layer interfaces	Solar cells; detector-relevant perovskite films	Extraction/loss competition where carrier collection or non-radiative loss can be diagnostically linked to the 2D component	PL/TRPL change, charge-collection evidence, trap/recombination signatures, photovoltaic or detector endpoint	ref. 17, 20, 21, 22

2.1 Metallic and electrode-facing interfaces: graphene and MXenes

Graphene-based interfaces mostly fall into three electrode-facing roles, as exemplified by the graphene/perovskite contact shown in Fig. 2a.⁸ In transparent-contact geometries, laminated graphene top electrodes provide optical transmission and lateral conduction in semitransparent perovskite solar cells, while graphene-containing GRAPE stacks extend this contact strategy to scalable panel architectures.^{6,15} In electronically coupled contacts, molecularly thin 2D Ruddlesden–Popper perovskite/graphene van der Waals interfaces show a lower charge-injection barrier than Au and femtosecond photoinduced charge transfer, identifying graphene as an extraction or tunnelling contact.⁸ In electrode-stabilizing architectures, Cu–Ni/graphene rear electrodes use bifacial graphene as a diffusion barrier against oxygen/moisture ingress and parasitic metal/device reactions, while the Cu–Ni alloy sets the electrode work function.⁴ These graphene examples share a geometry-defined interface logic: the same sp^2 -carbon sheet changes function mainly through where it is placed and how continuous the contact is, rather than through intrinsic surface-termination chemistry.^{4,6,8,15}

MXenes define a termination-controlled metallic interface motif at the perovskite/ETL contact, as illustrated by the band-profile comparison in Fig. 2b.³ In $Ti_3C_2T_x$ -modified perovskite solar cells, the same metallic carbide sheet is used to tune both sides of the interface: surface terminations shift the work function of the perovskite absorber, and the same termination chemistry shifts the work function of the TiO_2 electron-transport layer. This dual work-function tuning modifies the perovskite/ TiO_2 band alignment, reduces the interfacial extraction barrier, and improves charge extraction at the selective contact. Compared with graphene, whose role is mainly set by contact geometry and film continuity, $Ti_3C_2T_x$ is defined by termination-dependent electrostatic control at the perovskite/ETL interface.³

2.2 Semiconductor/perovskite heterostructures: TMDs and black phosphorus

TMD/perovskite interfaces extend beyond electrode-facing contact engineering when the 2D crystal contributes phase-controlled extraction,¹¹ coupling-controlled optical gain,¹⁰ or spin/valley-selective excitonic response.⁹ A representative coupling-sensitive perovskite/TMD heterostructure is shown in Fig. 2c.¹⁰ In solution-processed MoS_2 /perovskite photodetectors, MoS_2 controls carrier extraction through its electronic phase: metallic-phase MoS_2 improves transport, whereas finite-bandgap MoS_2 changes the extraction pathway and photodetector response.¹¹ When $MAPbI_3$ is deposited on MoS_2 flakes, the layered MoS_2 surface functions as a van der Waals epitaxial template, controlling perovskite nucleation and crystallinity while improving interfacial hole extraction.¹⁴ In facet-selective van der Waals growth of perovskite/TMD heterostructures, monolayer TMDs direct single-crystalline perovskite growth with controlled interfacial crystal planes, reducing energetic disorder at the mixed-dimensional interface and improving optical gain and lasing behaviour.¹⁰ In stacked chiral perovskite/monolayer MoS_2 heterostructures, chiral-induced spin filtering drives spin-selective charge transfer, changes the valley population of MoS_2 , and generates room-temperature valley polarization.⁹

Black phosphorus adds an anisotropic finite-bandgap branch to perovskite–2D heterostructures. Unlike gapless graphene contacts, few-layer BP provides direction-dependent electronic and optical response arising from high-mobility transport anisotropy and linear dichroism.¹² Waveguide-integrated BP photodetectors use its narrow but finite bandgap for near-infrared detection with low dark current, separating BP-type photodetection from gapless graphene contacts.¹³ In perovskite – 2D systems, this finite-bandgap and anisotropic character makes BP relevant to interfaces where charge extraction, recombination loss and photodetector response are coupled to direction-dependent carrier transport rather than to a metallic contact. BP quantum dots and BP-derived interfaces have further been used to modify perovskite or transport-layer interfaces, linking BP-containing systems to hole extraction, interfacial recombination modulation and photovoltaic or detector endpoints.¹⁶ Accordingly, BP is best viewed here as an anisotropic finite-bandgap interface family, while direct perovskite/BP excitonic-coupling examples remain less established than TMD/perovskite systems.

2.3 Structure- and field-mediating interfaces

Structure- and field-mediating interfaces are defined by the dominant variable controlled by the 2D component, not by the material name alone. A representative buried field-mediated interface is shown in Fig. 2d.¹⁷ MoS₂-assisted solution-phase epitaxy uses MoS₂ flakes as buried growth substrates rather than phase-engineered transport layers; the layered van der Waals surface directs MAPbI₃ nucleation and crystallographic orientation, increasing crystallinity and reducing growth-related disorder before hole extraction is improved.¹⁴ Strain mediation is illustrated by WS₂/CsPbBr₃ van der Waals heterostructures, where WS₂ is inserted at the electron-transport-layer/perovskite boundary and its atomically smooth, dangling-bond-free surface releases tensile strain in CsPbBr₃, reducing strain-associated defects and improving device stability.¹⁸ Grain-boundary pathway control appears in BP-, MoS₂-, and graphene-oxide-flake-modified MAPbI₃ films: the flakes localize at grain-boundary edges and create local hole-conducting pathways from grain boundaries to the hole-transport layer, so the improvement arises from grain-boundary transport geometry rather than a continuous planar perovskite/2D heterojunction.⁷ Cl-terminated MXene buried interfaces provide a field-mediated route: the Cl-rich MXene surface introduces interfacial dipoles that bend the local perovskite bands and induce n-type character near the buried interface. The resulting nanoscale field-effect junction improves carrier extraction and suppresses recombination through electrostatic steering, rather than through MXene acting as a simple planar charge-accepting layer.¹⁷

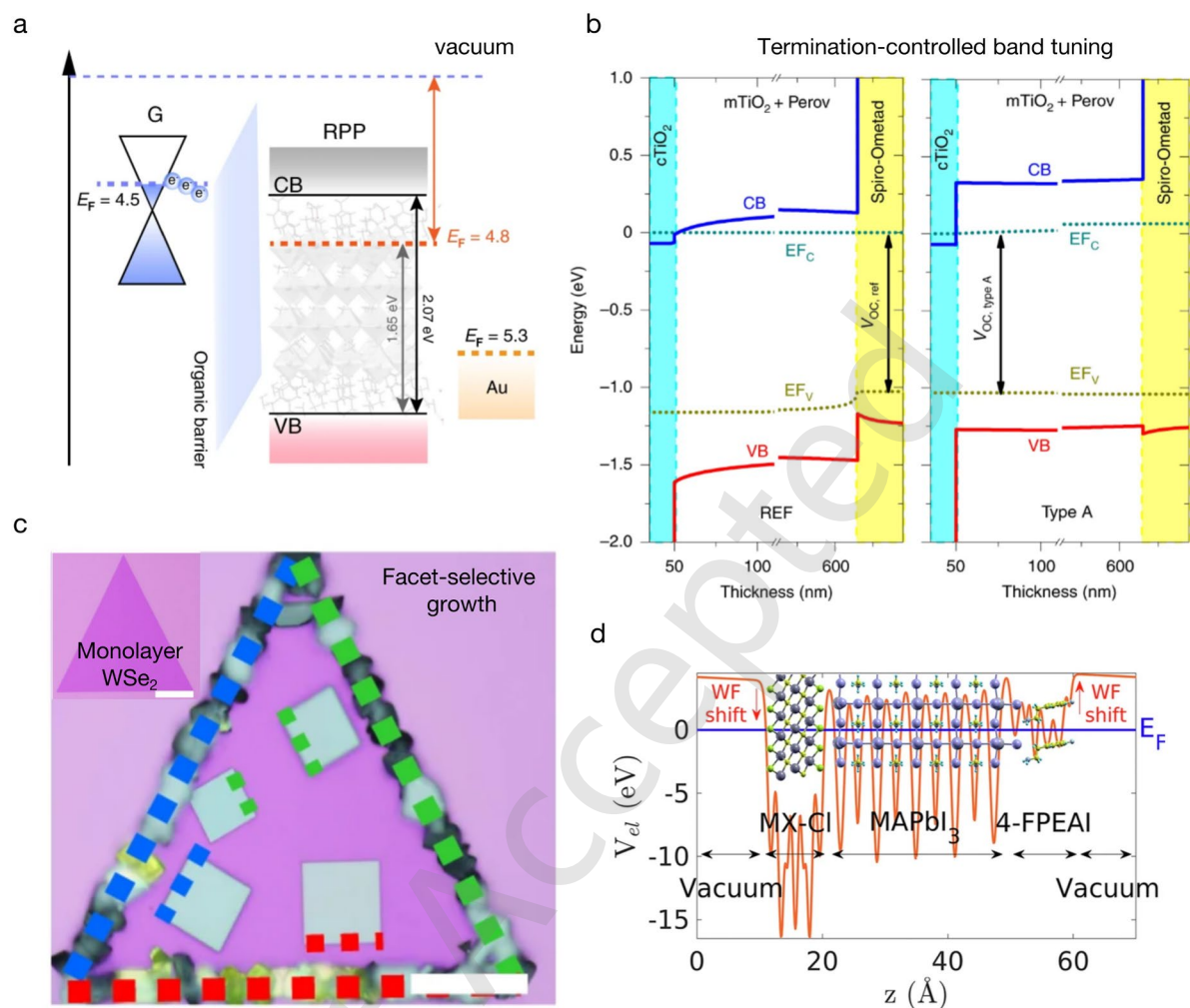
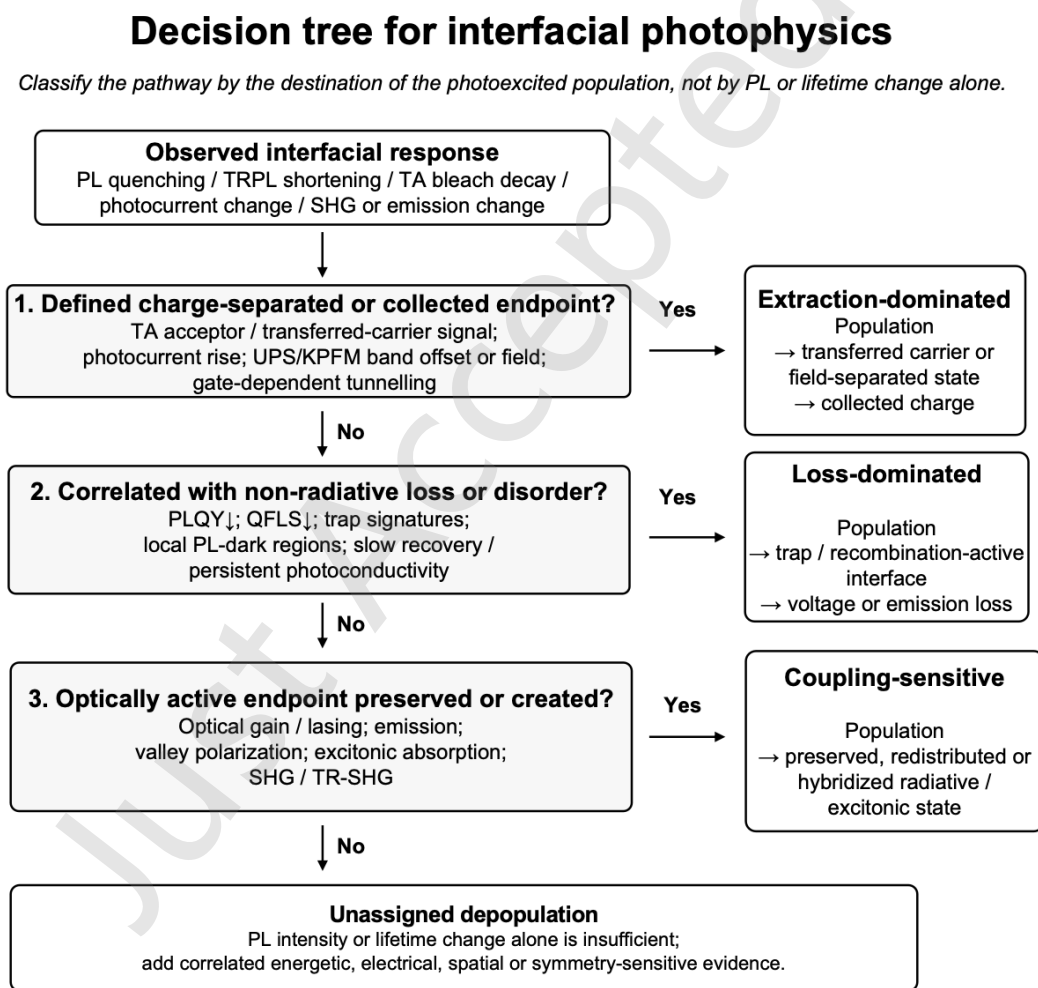


Figure 2 Representative interface archetypes in perovskite–2D heterostructures. a, Electrode-facing graphene contact, illustrated by the interfacial energy alignment of a molecularly thin perovskite/graphene junction. b, Termination-controlled MXene contact, showing work-function tuning and band-profile modulation at selective interfaces. c, Coupling-sensitive perovskite/TMD heterostructure, represented by facet-selective growth on a monolayer TMD template. d, Field-mediated buried interface, illustrated by the electrostatic potential profile of a Cl-terminated MXene/perovskite junction. Panel a adapted under the CC BY 4.0 license from Ref. 8 ©2020, The Author(s). Panel b adapted with permission from Ref. 3 ©2019, Springer Nature. Panel c adapted under the CC BY 4.0 license from Ref. 10 ©2024, The Author(s). Panel d adapted under the CC BY 4.0 license from Ref. 17 ©2026, The Author(s).

3. Interfacial photophysics and its spectroscopic readout

Interfacial photophysics in perovskite-2D heterostructures is governed by the redistribution of photoexcited states among charge-separated, non-radiative, and coupled radiative pathways. A perovskite excitation may be converted into spatially separated carriers when the interface provides

a tunnelling pathway, a favourable band offset, or a local electric field. It may be lost when the interface introduces traps, energetic disorder, or recombination-active grain-boundary and surface regions. It may also be preserved or redistributed into optically active coupled states when the interface supports selective transfer, interfacial excitonic response, or radiative-state management. These pathways can produce similar first-order observables, especially PL quenching or lifetime shortening, so the physical assignment must follow the destination of the excited population rather than the change in lifetime alone. Before discussing these regimes in detail, Box 1 summarizes a diagnostic decision tree that assigns observed interfacial responses according to the destination of the photoexcited population and the supporting experimental evidence.



Box 1. Diagnostic decision tree for interfacial photophysics. A schematic workflow for translating common interfacial observables into mechanism assignments. The tree highlights that reliable classification requires endpoint-specific, correlated evidence rather than relying on PL quenching or lifetime changes alone.

3.1 Extraction- and separation-dominated interfaces

Extraction- and separation-dominated interfaces convert a perovskite-localized photoexcited population into an interfacial charge-separated state. After optical excitation, the perovskite hosts

an electron–hole population in the absorber or quantum-well-like region. An extraction-active interface changes the boundary condition of this population: one carrier accesses a lower-energy or more strongly coupled final state in an adjacent 2D layer, electrode, or transport contact, while the opposite carrier remains in the perovskite or exits through a complementary pathway. The essential photophysical step is therefore not faster decay, but reduced electron–hole spatial overlap through tunnelling, band-offset-driven transfer, or field-driven separation before radiative or trap-assisted recombination occurs, as illustrated for a tunnelling-enabled perovskite/graphene junction in Fig. 3a.^{3,8,11,17,19,20}

In molecularly thin 2D Ruddlesden–Popper perovskite/graphene contacts, separation is governed by wavefunction access across a van der Waals tunnelling barrier. The perovskite supplies the optically excited population, while graphene supplies a continuum-like conducting final state with a lower injection barrier than Au. The plane-contacted geometry reduces the effective tunnelling distance, enabling photoinduced charge transfer on the femtosecond timescale. Electrical measurements further show a gate-tunable transition from direct tunnelling to field emission, so the optical depopulation is tied to an interfacial tunnelling channel rather than to generic quenching.⁸ This combination of barrier reduction, femtosecond transfer, and gate-dependent tunnelling connects the optical signal to carrier injection across the van der Waals contact.⁸

Band-offset-driven charge separation in TMD heterobilayers provides a well-established conceptual reference for this separation logic. In MoS₂/WS₂ heterobilayers, excitation of the MoS₂ A exciton initially creates an intralayer excitonic population. This example is therefore provided as a conceptual reference for establishing the spectroscopic criteria needed to assign ultrafast charge separation in perovskite–2D interfaces. The valence-band alignment favours hole transfer into WS₂, while the electron remains in MoS₂, converting an intralayer exciton into a spatially indirect electron–hole population across the interface within about 50 fs.¹⁹ Ultrafast studies of van der Waals heterostructures show that this conversion depends on band alignment, interlayer coupling, dielectric environment, and excitonic structure, so a fast decay can be assigned to charge separation only when donor- and acceptor-layer populations are spectrally resolved.^{19,20}

Work-function-engineered and field-mediated perovskite–2D contacts provide extraction without requiring a conventional planar semiconductor heterojunction. In Ti₃C₂T_x-modified perovskite/TiO₂ contacts, MXene terminations shift the work functions of both the perovskite absorber and the TiO₂ electron-transport layer, reshaping the perovskite/ETL band alignment and lowering the barrier for electron extraction.³ The physical effect is to bias the photoexcited carrier distribution toward the selective contact before interfacial recombination returns carriers to the perovskite. Cl-terminated MXene buried interfaces add an electrostatic route: termination-induced dipoles bend the local perovskite bands and induce n-type character near the buried interface, creating a nanoscale field-effect junction that separates carriers through local electric-field steering rather than through MXene acting as a simple planar acceptor.¹⁷

The spectroscopic readout of extraction must capture population conversion, not only population decay. TRPL shortening or PL quenching marks the disappearance of perovskite emission, but it

does not specify whether the population enters graphene, a state of TMD, a transport layer, or a trap. In the perovskite/graphene case, the lower injection barrier, femtosecond photoinduced transfer, and gate-dependent tunnelling behaviour connect optical depopulation to carrier injection.⁸ In MoS₂/WS₂, femtosecond pump–probe spectra and PL mapping distinguish the heterostructure response from isolated monolayers and assign the ultrafast signal to hole transfer from MoS₂ to WS₂.¹⁹ A perovskite bleach decay should therefore be treated as extraction only when it is paired with a material-specific endpoint: a graphene tunnelling channel, a TMD-layer population response, a field-separated interfacial state, or photocurrent arising from separated carriers.^{3,8,11,17,19,20}

Energetic and electrical measurements close the assignment. UPS, Kelvin probe, or KPFM support extraction when they show work-function shifts, band offsets, or dipole-induced band bending that create a downhill separation pathway.^{3,17} TA supports extraction when donor-state decay is accompanied by formation of a transferred-carrier or charge-separated-state signal rather than by simple loss of oscillator strength.^{8,19,20} Photocurrent dynamics support extraction when the separated population survives long enough to appear as collected current, as in phase-dependent MoS₂/perovskite photodetectors where the MoS₂ electronic phase changes carrier collection and device response.¹¹

Thus, extraction-dominated photophysics requires the full chain: photoexcited perovskite population → interfacial driving force → tunnelling, transfer, or field-driven separation → spatially separated carrier population → collected charge. Fast decay is assigned to extraction only when such an endpoint is experimentally resolved.

3.2 Loss-dominated and disorder-sensitive interfaces

Loss-dominated interfaces convert photoexcited states into heat-producing non-radiative decay rather than useful charge or radiative output. The initial excitation may still be created in the perovskite absorber, but the interfacial landscape redirects the carrier population into trap states, recombination-active surfaces, grain-boundary disorder, or defective buried contacts. The key physical step is capture into a non-radiative configuration: electron–hole overlap is restored at a defect, multiphonon relaxation removes the excitation energy, and the device loses quasi-Fermi-level splitting, photocurrent quality, or emission yield.^{21–25}

In solar-cell interfaces, this loss pathway appears as a reduction of quasi-Fermi-level splitting rather than merely a shorter optical lifetime. The QFLS-based separation of interfacial and bulk recombination shown in Fig. 3c can be provided as a methodological reference for diagnosing interfacial non-radiative loss. Absolute and transient PL imaging of planar perovskite solar cells has visualized interfacial recombination at transport-layer contacts and connected the spatial PL loss to open-circuit-voltage limitations.²¹ Multidimensional PL imaging of efficient inverted perovskite solar cells further converts local emission deficits into quantitative maps of non-radiative loss at buried interfaces.²² These measurements show that a dark interface is not automatically an extraction interface: if the removed excitation does not produce separated carriers, the lost PL represents non-radiative recombination.

Loss can also be local and dynamic. Temperature-dependent luminescence blinking reveals microscopic non-radiative decay sites in perovskites that switch between emissive and quenching states.²³ Operando trapped-carrier measurements show that trap populations under working conditions are linked to non-radiative recombination, making loss a device-operating-state property rather than a static material label.²⁴ This matters for perovskite–2D interfaces because a 2D layer can either bypass local loss pathways or introduce new ones, depending on where it sits, how clean the contact is, and whether the interface adds energetic disorder.

Grain-boundary-localized 2D flakes illustrate the distinction between useful local routing and non-radiative loss. BP, MoS₂, and graphene oxide flakes positioned at perovskite grain-boundary edges can form local hole-conducting pathways toward the hole-transport layer.⁷ The relevant photophysics is not a uniform planar perovskite-to-2D transfer event, but a local change in the competition between grain-boundary recombination and hole removal. If the flake pathway removes holes before they recombine at the boundary, the interface suppresses loss; if the added interface introduces trap-rich contacts, the same geometry can quench PL. The physical assignment therefore depends on whether local carrier routing reduces or increases the probability of non-radiative capture.^{7,21–25}

PL quenching or TRPL shortening is assigned to loss only when depopulation is correlated with lower PLQY, local PL darkening, QFLS loss, slow recovery, persistent photoconductivity, trap signatures, or the absence of a transferred-carrier or photocurrent endpoint. The difference is the destination of the population, not the sign of the lifetime change.^{3,8,11,17,19,21–25}

Spatial correlation is the strongest way to assign disorder-sensitive loss. A local PL-dark region becomes a non-radiative site when it overlaps with surface-potential disorder, recombination-active grain boundaries, or voltage-loss maps.^{21,22} A buried interface becomes a loss channel when its PL loss is accompanied by reduced quasi-Fermi-level splitting rather than increased collected current.^{21,22} A trap-rich photodetector interface may increase gain through photogating, but that gain is usually accompanied by dark-current penalties, slow response recovery, or persistent carriers.^{7,11,24} The same interface can therefore be beneficial for one device metric and detrimental for another.

Loss-dominated photophysics is the pathway photoexcited population → trap or recombination-active interface → non-radiative relaxation → voltage, current-quality, or emission loss. This pathway is not the absence of interfacial activity; it is an active conversion of optical excitation into non-radiative decay. Its spectroscopic signature is not simply weak PL, but weak PL tied to traps, local disorder, voltage loss, or dark-current-related carrier storage.

3.3 Coupling-sensitive and radiative-state-preserving interfaces

Coupling-sensitive and radiative-state-preserving interfaces support optical functionality through two related but mechanistically distinct routes. In a structure-mediated radiative-state-preserving interface, the 2D component improves crystallinity, reduces energetic disorder or suppresses non-radiative pathways, thereby preserving the optically active population without necessarily

modifying the interfacial electronic or excitonic states directly. In a direct interfacial-coupling route, the interface modifies excitonic, spin, valley or electronic degrees of freedom and produces corresponding state-specific optical responses. This regime is most relevant when the device output depends on emission, optical gain, lasing, valley polarization, or nonlinear optical response. The interface must then manage the excited-state population without destroying the radiative or excitonic character needed for optical function. Strong extraction can be useful for a photodetector, but the same depletion can suppress gain or emission if it removes carriers from optically active states.

Facet-selective perovskite/TMD heterostructures illustrate the structure-mediated radiative-state-preserving route, in which improved crystallinity, reduced energetic disorder and suppressed non-radiative scattering preserve the optically active population and enhance optical gain and lasing without requiring direct modification of an excitonic or electronic state (Fig. 3b). Monolayer TMDs direct single-crystalline perovskite growth with controlled interfacial crystal planes, reduce energetic disorder at the mixed-dimensional interface, and improve optical gain and lasing behaviour.¹⁰ The photophysical effect is not simply carrier removal from the perovskite. A cleaner and more ordered interface reduces inhomogeneous energy landscapes and non-radiative scattering pathways, allowing a radiative population to survive long enough to support gain and lasing, as reflected by the pump-dependent lasing evolution in Fig. 3d.¹⁰ In this regime, the useful interface is not the fastest quencher, but the one that controls coupling while preserving an optically active carrier distribution.

By contrast, chiral perovskite/monolayer MoS₂ heterostructures illustrate a direct interfacial-coupling route. Chiral-induced spin filtering drives spin-selective charge transfer from the perovskite to MoS₂, modifies the valley population of monolayer MoS₂, and produces room-temperature valley polarization.⁹ The endpoint is a spin-filtered, polarization-resolved excitonic population in the 2D semiconductor, rather than only separated charge.⁹ Accordingly, improved emission, optical gain or lasing alone is treated as evidence of radiative-state preservation rather than direct interfacial coupling, unless it is accompanied by state-specific signatures such as layer-resolved population transfer, excitonic resonances, polarization- or valley-resolved responses, or hybridized-state features.

This regime can also be considered in relation to interlayer-exciton physics in TMD van der Waals heterostructures, which provides a conceptual framework for identifying coupling-sensitive behaviour in perovskite–2D interfaces. In TMD van der Waals heterostructures, interlayer excitons form when electrons and holes occupy different layers while remaining Coulomb-bound across the interface. Their lifetime, oscillator strength, diffusion, and emission depend on the balance between charge separation and electron–hole overlap.²⁶ Optical absorption measurements of interlayer excitons show that weak PL does not necessarily imply weak excitonic oscillator strength, because absorption can reveal optically active interlayer excitons that are not bright in emission.²⁷ For perovskite/TMD and chiral perovskite/MoS₂ interfaces, PL intensity, lifetime, transfer rate, optical gain, absorption, and polarization response are therefore different projections of the same coupled excited-state landscape.^{9,10,26,27}

Coupling also changes the meaning of ultrafast dynamics. Excitonic interactions can mediate ultrafast interlayer photoexcited response in van der Waals heterostructures, so a fast spectral change is not always a simple single-particle electron-transfer event.²⁸ Perovskite–2D interfaces are especially susceptible to this ambiguity because free carriers, excitons, polarons, trapped carriers, and radiative states can overlap in energy and time. A coupling-sensitive assignment therefore requires evidence that the interface creates, preserves, or redistributes an optically active state.

Radiative-state preservation gives a different interpretation of PL. In extraction-dominated contacts, PL quenching can be desirable if it reflects carrier collection.^{8,11,17} In gain or emission interfaces, excessive PL quenching can be detrimental because it depletes the carrier population required for stimulated emission.¹⁰ In spin/valley interfaces, PL polarization or valley contrast becomes more important than total PL intensity because the interface transfers information into an excitonic degree of freedom.⁹ In interlayer-exciton systems, absorption and polarization can reveal coupling even when emission is weak.^{26,27} The useful observable therefore depends on the intended photophysical output.

SHG provides a complementary readout for coupling-sensitive interfaces because it probes symmetry, polarity and field-sensitive optical response. The examples discussed below, including the electrically modulated and polarization-dependent SHG responses shown in Fig. 3e and f, are drawn from related low-dimensional systems and are provided as methodological references for perovskite–2D interfaces. In layered non-centrosymmetric materials, SHG can identify crystal symmetry and second-order nonlinear response, as shown in 2D MnTeMoO₆ and its polarization-dependent SHG response (Fig. 3f).²⁹ Related perovskite systems provide useful methodological analogies: pressure-dependent SHG reveals hidden non-centrosymmetry in globally centrosymmetric 2D halide perovskites,³⁰ while rotational SHG resolves interfacial symmetry breaking in oxide heterostructures.³¹ These examples show that SHG can distinguish whether a nonlinear optical signal originates from intrinsic lattice symmetry, mechanically induced symmetry breaking or an interface-specific polar environment.

Beyond symmetry identification, SHG can also be modulated by electrostatic doping, applied electric fields and ferroelectric polarization. Gate-controlled monolayer WSe₂ shows that the second-order nonlinear response of a 2D semiconductor can be tuned near excitonic resonances, providing a representative example of electrically modulated SHG readout (Fig. 3e).³² Electric-field-induced SHG enhancement in 2D hybrid perovskites further demonstrates that resonant nonlinear response can be activated or amplified by an external field.³³ Ferroelectric/TMD heterointerfaces extend this principle to interfacial polar coupling, where ferroelectric domains can enhance, suppress or reorient nonlinear optical response in the adjacent 2D layer.^{34,35} Although several of these examples come from 2D halide perovskites, oxide heterostructures or ferroelectric/TMD interfaces rather than the primary halide perovskite/non-perovskite 2D-material systems discussed here, they define a clear design rule for future perovskite–2D studies: symmetry breaking should be measured directly, and its field- or polarity-dependent tunability should be tested dynamically.

Time-resolved SHG would extend this symmetry sensitivity into the excited-state regime. Static SHG maps broken symmetry, crystal orientation, or polar order, but it does not reveal how photoexcitation changes interfacial polarity, transient fields, or coupled radiative states. Time-resolved SHG could track pump-induced charge separation, transient interfacial dipoles, symmetry changes, and exciton-related nonlinear response on the same timescale as TA and TRPL. This is especially relevant for mixed-dimensional perovskite-2D interfaces, where extraction, loss, and coupling can all occur within ultrafast or early-time carrier-relaxation windows.^{8–10,17,19,20,32}

Coupling-sensitive photophysics follows the pathway photoexcited population → selective transfer or interfacial coupling → preserved, redistributed, or hybridized radiative state → optical gain, emission, valley polarization, or nonlinear optical response. The spectroscopic signature is not merely slower or faster decay, but an optically active endpoint.

3.4 Spectroscopic logic: from observables to mechanisms

Spectroscopic assignment requires matching each observable to the physical quantity it measures. TRPL measures the survival of the emissive population in the perovskite, but not its destination. A faster decay therefore only establishes depopulation of the radiative state; extraction is supported when this decay is accompanied by transferred-carrier formation or photocurrent, whereas loss is supported when it is accompanied by reduced PLQY, quasi-Fermi-level-splitting loss, or spatially localized dark regions.^{8,11,21–25}

TA tracks population redistribution across spectrally distinct states. A donor bleach decay paired with growth of a layer-specific carrier signal or a long-lived charge-separated feature supports interfacial separation.^{8,19,36} Decay of the perovskite signal without a new carrier population points instead to non-radiative depopulation when correlated with traps or voltage loss.^{23–27} Spectral evolution involving excitonic resonances, valley response, or gain-related states indicates redistribution into coupled radiative or excitonic pathways.^{9,10,26–28}

UPS, Kelvin probe, and KPFM measure the energetic and electrostatic landscape that determines whether the observed dynamics are physically allowed. Work-function shifts, band offsets, and interface dipoles support extraction or field-driven separation when they create a downhill pathway for one carrier.^{3,17} Local surface-potential disorder supports loss when it coincides with PL-dark regions or recombination-active interfaces.^{21,22} Photocurrent dynamics test whether separated carriers survive as useful current. A fast optical decay with a corresponding photocurrent rise supports extraction, whereas slow recovery, persistent photoconductivity, or dark-current increase indicates trapping or photogating.^{7,11,24}

SHG and time-resolved SHG provide a separate symmetry-sensitive readout. Conventional TRPL and TA track population decay and spectral redistribution, but they do not directly resolve inversion-symmetry breaking, polar order, crystal orientation, or transient interfacial fields. Static SHG can identify non-centrosymmetric order and field-sensitive nonlinear response, while TR-SHG could follow photoinduced symmetry modulation, transient polar fields, and interfacial electric-field formation on ultrafast timescales. This makes SHG/TR-SHG particularly relevant for coupling-

sensitive and field-mediated perovskite–2D interfaces, where the key photophysics may involve photoinduced symmetry breaking or transient interfacial polarity rather than only carrier extraction or recombination.^{29,32}

A robust mechanism assignment therefore requires population-sensitive, energetic, electrical, spatial, and symmetry-sensitive observables to converge on the same excited-state pathway: charge separation, non-radiative loss, or coupled radiative response. Table 2 translates this requirement into observable-specific criteria for distinguishing extraction, recombination loss, trapping, interfacial coupling and radiative-state preservation.

Table 2 | Observable-specific criteria for assigning interfacial photophysical pathways.

Observable	What it measures	Evidence supporting extraction	Evidence supporting recombination loss or trapping	Evidence supporting interfacial coupling or radiative-state preservation	Quantitative descriptors to report	Refs.
TRPL	Survival of the emissive perovskite population; does not identify the population endpoint by itself.	Lifetime shortening assigned to extraction only when paired with transferred-carrier formation, photocurrent generation or a downhill energetic pathway.	Shorter lifetime with lower PLQY/QFLS, local PL-dark regions, trap signatures, slow recovery or persistent carriers indicates non-radiative depopulation or trapping.	Preserved or optimized radiative lifetime together with high PLQY, emission, gain or polarization-resolved response supports radiative-state preservation.	Lifetime components, amplitudes, fluence dependence, temperature dependence, transfer/quenching time constants.	refs. 8, 10, 21 – 25
TA	Population redistribution among spectrally resolved donor, acceptor, charge-separated, trap and excitonic states.	Donor bleach decay accompanied by acceptor-layer signal, transferred-carrier feature or long-lived charge-separated population; femtosecond transfer examples should be assigned to a final state.	Perovskite signal decay without a new carrier population, or with trap-state absorption / long-lived dark-carrier signatures, supports loss or trapping.	Excitonic resonances, valley-selective response, gain-related stimulated-emission features or hybridized-state signatures support coupling-sensitive behaviour.	Rise/decay constants, transfer time, spectral assignment, charge-separated lifetime, excitation-density dependence.	refs. 8, 19, 20, 26 – 28, 36
PLQY / absolute PL	Balance between radiative and non-radiative recombination under defined excitation and stack geometry.	PL quenching can indicate extraction only when accompanied by collected charge, photocurrent or an independently verified extraction pathway.	PLQY decrease, spatial darkening or increased non-radiative rate without a charge-collection endpoint indicates recombination loss.	Maintained or increased PLQY, emission narrowing, amplified spontaneous emission or lasing supports radiative-state preservation in emission/gain contexts.	Absolute PLQY, radiative/non-radiative rates, intensity maps, linewidth, excitation-density dependence.	refs. 9, 10, 21 – 23, 25 – 27
QFLS	Thermodynamic free-energy splitting of photogenerated carriers and voltage loss associated with recombination.	Extraction-compatible interfaces should preserve QFLS while improving collection; QFLS alone does not prove extraction.	Reduced QFLS, local QFLS heterogeneity or interface-specific QFLS loss directly indicates non-radiative recombination.	High QFLS together with strong emission or gain supports preservation of radiative carrier populations.	QFLS value, Δ QFLS, implied Voc, non-radiative voltage loss, spatial distribution.	refs. 21, 22, 25
UPS / Kelvin probe / KPFM	Work function, band offsets, interfacial dipoles, band bending and local surface-potential disorder.	Work-function shift, favourable band offset, barrier reduction or dipole-induced field that provides a downhill separation pathway.	Potential disorder, unfavourable alignment, carrier accumulation, charged-defect potentials or local fields correlated with PL loss indicate recombination/trapping risk.	Interfacial polarity or field environment that modulates excitonic, nonlinear or valley response supports coupling only when paired with optical evidence.	Work-function shift, band offset, contact-potential difference, band bending, field strength, potential-disorder width.	refs. 3, 8, 17, 21, 22, 32 – 35

Observable	What it measures	Evidence supporting extraction	Evidence supporting recombination loss or trapping	Evidence supporting interfacial coupling or radiative-state preservation	Quantitative descriptors to report	Refs.
Photocurrent dynamics	Whether separated carriers survive as useful current or remain stored as trapped / photogating charge.	Fast photocurrent rise synchronized with optical depopulation, efficient collection, low dark current and limited hysteresis support extraction.	Slow rise/decay, persistent photoconductivity, increased dark current, long recovery or photogating-dominated gain indicate trapping or carrier storage.	For emission/gain devices, photocurrent can reveal destructive extraction; coupling-sensitive assignment requires accompanying optical endpoint.	Rise/fall times, bandwidth, responsivity, detectivity, dark current, linear dynamic range, hysteresis, bias dependence.	refs. 7, 11, 37 – 39
SHG / TR-SHG	Broken symmetry, polarity, transient interfacial fields and nonlinear/excitonic response.	Pump-induced SHG assigned to extraction only when correlated with transient charge separation, interfacial dipoles, TA or photocurrent endpoints.	Long-lived electric-field-induced SHG, persistent polarity changes or disorder-linked SHG contrast can indicate trapped charges or field-related loss.	Symmetry breaking, resonant excitonic SHG, valley/polar optical response or transient nonlinear signal supports interfacial coupling and radiative-state preservation.	SHG anisotropy, phase/polarization dependence, TR-SHG rise/decay time, field dependence, resonance energy, nonlinear coefficient where available.	refs. 29 – 35

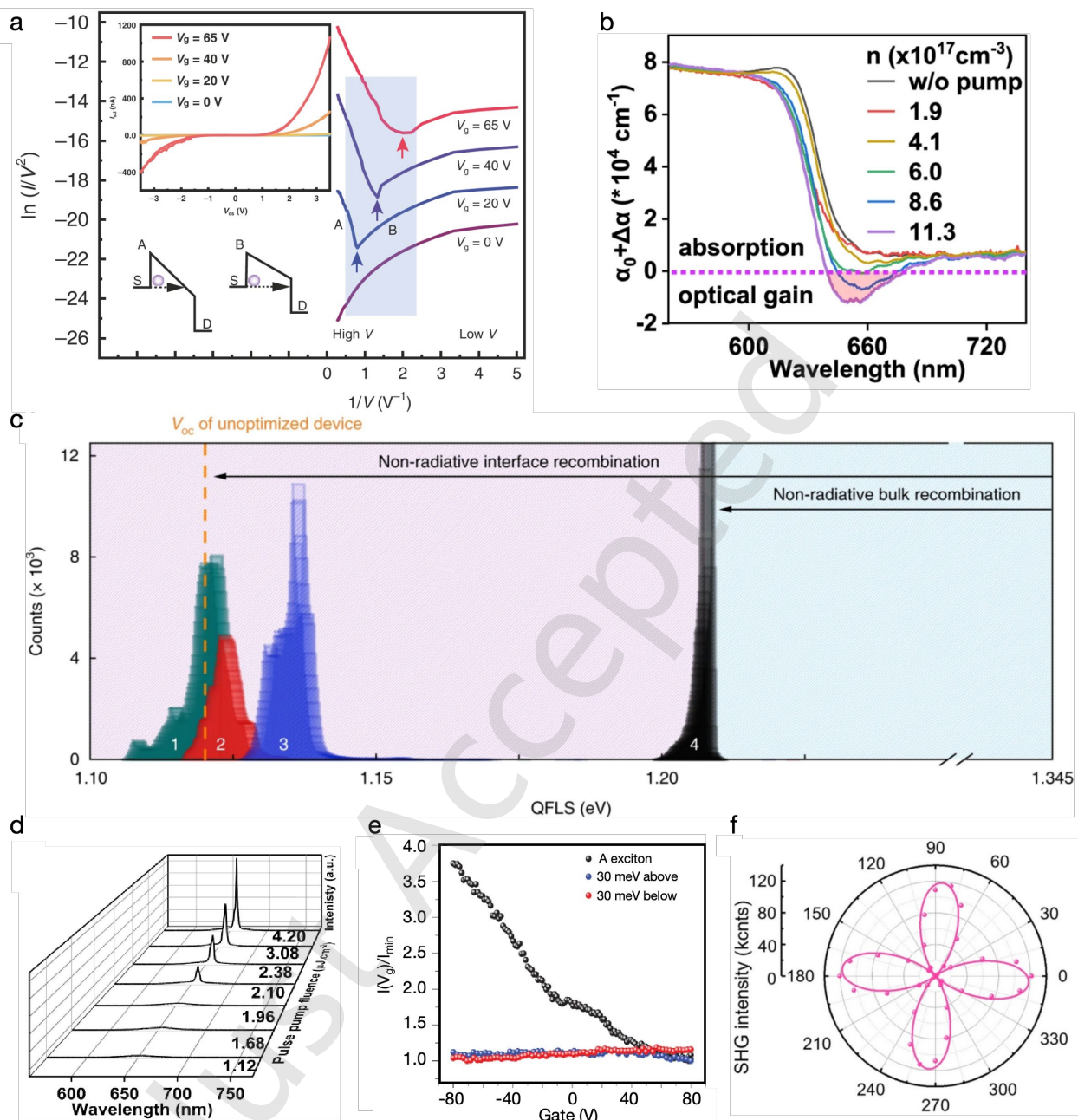


Figure 3 Interfacial photophysics and spectroscopic readouts relevant to perovskite-2D heterostructures. **a**, Tunnelling-enabled extraction pathway at a molecularly thin perovskite/graphene junction, showing the gate-dependent transition between direct tunnelling and Fowler–Nordheim tunnelling. **b**, Coupling/radiative pathway manifested as optical gain in a facet-selective perovskite/TMD heterostructure. **c**, Loss pathway dominated by non-radiative recombination, where the QFLS distribution distinguishes interfacial recombination from bulk recombination. **d**, Lasing evolution under increasing pump fluence, showing the transition from broad spontaneous emission to a sharp lasing mode. **e**, Gate-controlled SHG response, illustrating symmetry-sensitive nonlinear optical readout modulated by electrostatic doping. **f**, Polarization-dependent SHG from a non-centrosymmetric 2D crystal, highlighting strong symmetry-sensitive nonlinear optical response. Panel a adapted under the CC BY 4.0 license from Ref. 8 ©2020, The

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4. Device-level impact of interfacial photophysics

The device-level impact of a perovskite–2D interface is determined by the photoexcited-state pathway that dominates under operation. Solar cells benefit when the interface combines selective charge collection with suppressed non-radiative recombination and improved contact stability.^{3,4,6} Photodetectors expose a sharper competition: the same interface may accelerate carrier extraction or amplify the signal through trapped-carrier photogating, and the balance is reflected in responsivity, detectivity, response speed, and dark current.^{11,37–39} Light-emitting, gain, and lasing devices follow a different design logic, where maximal carrier removal can be detrimental and the useful interface instead preserves or redistributes radiative excited states.^{9,10}

4.1 Solar cells: extraction, recombination control, and stability

In perovskite solar cells, extraction-dominated and loss-suppressed interfaces improve performance through different but coupled routes. Charge-collection interfaces primarily affect current extraction and fill factor by reducing the barrier for one carrier to reach its selective contact before recombination occurs. $\text{Ti}_3\text{C}_2\text{T}_x$ MXene interface engineering illustrates this clearly: MXene terminations shift the work functions of the perovskite absorber and TiO_2 electron-transport layer, improve the perovskite/ETL band alignment, and thereby accelerate electron extraction.³ The device-level impact is not only higher photocurrent collection but also reduced hysteresis and improved fill factor, because better contact energetics suppress carrier accumulation and interfacial back transfer.³ By contrast, loss-suppressed interfaces act most directly on open-circuit voltage, V_{oc} . When the interfacial density of recombination-active states is reduced, a larger quasi-Fermi-level splitting can be maintained under illumination, so V_{oc} rises even if the optical absorption is unchanged.^{21,22,25} The practical distinction is that extraction-dominated interfaces improve collection efficiency, whereas loss-suppressed interfaces preserve the free energy of the photogenerated carrier population. Representative device-level manifestations of these interface regimes are summarized in Fig. 4.

For PSCs, these two routes often need to operate together. A selective contact that extracts carriers rapidly but introduces interfacial recombination will sacrifice V_{oc} and sometimes FF; a chemically benign interface that suppresses recombination but leaves a poor energetic alignment will limit charge collection.^{3,21,22} This is why interfacial photophysics translates directly into the coupled PSC metrics of V_{oc} and FF rather than into a single “better interface” label. V_{oc} is especially sensitive to non-radiative interfacial loss when the device fails to realize the quasi-Fermi-level splitting available in the absorber,²⁵ whereas FF is especially sensitive to contact selectivity, carrier accumulation, and transport bottlenecks near the interface.^{3,21,22}

Stability adds a third consequence that is not captured by early-time carrier dynamics alone, but rather important especially for perovskite-based devices. Graphene-protected Cu–Ni rear electrodes show that an electrode-facing 2D interface can improve device lifetime by suppressing oxygen and moisture ingress and by blocking parasitic chemical reactions between the metal electrode and neighbouring layers.⁴ Here, the beneficial device-level impact arises from a combined photophysical and chemical effect: more robust interfacial energetics under operation, reduced degradation-induced recombination, and preserved contact function during light, humidity, and thermal stress.⁴ The corresponding operational-stability advantage is shown in Fig. 4c, where CNG-based devices retain normalized efficiency far longer than conventional metal-electrode controls. At the module scale, graphene-containing GRAPE-type perovskite solar panels show that such interfacial design is not restricted to small-area cells; the same logic can support scalable operation and field-relevant stability.⁶ The corresponding extension from cell-scale interface engineering to outdoor photovoltaic deployment is illustrated in Fig. 4b. Solar-cell interfaces therefore reward a specific combination of interfacial functions: selective extraction, recombination suppression, and chemically stable contact formation.^{3,4,6}

4.2 Photodetectors: gain, speed, and dark current

Photodetectors expose interfacial photophysics more sharply than solar cells because their key metrics respond in opposite directions to extraction and trapping. A clean extraction interface tends to reduce carrier transit time, speed up response, and keep dark current under control if the contact remains selective.^{11,37,39} A trap-amplifying interface can produce very high responsivity and detectivity by photogating, but it often does so by storing one carrier species and letting the other recirculate through a high-mobility channel, which slows response and can increase dark current or persistent photocurrent.^{37,39} The central trade-off is therefore mechanistic: high responsivity can originate from efficient photocarrier conversion or from long-lived trapped carriers, and only the accompanying speed and dark-current behaviour distinguishes the two cases.^{37–39}

Perovskite–graphene hybrid photodetectors are a representative photogating case. Photogenerated carriers in the perovskite modulate the conductance of the graphene channel, so the interface converts optical excitation into a gate-like electrostatic perturbation of a high-mobility 2D conductor.³⁸ This mechanism can produce very large responsivity because one trapped carrier population can control many channel carriers, but the gain is tied to trapped-carrier lifetime and therefore tends to slow the photoresponse.³⁸ The device consequence is a familiar responsivity–speed tension: gain increases when carrier storage is long lived, whereas fast response requires those stored carriers to be released or removed quickly. A representative device-level readout of this photogating regime is shown in Fig. 4d&e, where the perovskite–graphene hybrid detector exhibits broadband photocurrent, high photoresponsivity and high EQE.

Perovskite–TMD photodetectors span a broader range between extraction and trapping. In hybrid perovskite/TMD devices, band-offset-driven transfer into a semiconducting 2D channel can produce efficient photoresponse with better channel control than a purely photogated graphene device, but the same interface can still become trap dominated if it introduces slow carrier release or leakage

pathways.³⁹ This makes dark current and recovery dynamics unusually informative. A nominally high-gain detector is not necessarily a good interface if the gain is obtained by interfacial carrier storage that raises dark current or prolongs recovery.^{37,39} Recent hybrid perovskite/MoS₂ photodetectors show that careful interface design can partially break the conventional responsivity–speed trade-off, indicating that high responsivity does not need to rely exclusively on slow trapping; efficient separation and controlled interfacial transport can raise signal while preserving faster response.³⁷ In photodetectors, more clearly than in solar cells, the device metrics themselves reveal which interface regime dominates: responsivity alone is ambiguous; responsivity together with speed and dark current identifies whether the interface is extraction-enhancing or trap-amplifying.

4.3 Light emission, optical gain, and lasing

Emission and gain devices invert the logic of extraction interfaces: charge transfer that improves carrier collection in solar cells or photodetectors can quench light emission or optical gain when it depletes the radiative-state population. The useful interface is therefore not the one that extracts most aggressively, but the one that preserves radiative states while still providing controlled interfacial coupling.^{9,10}

Facet-selective perovskite/2D semiconductor heterostructures illustrate this point directly. Monolayer TMDs can direct single-crystalline perovskite growth with controlled interfacial crystal planes, reduce energetic disorder at the mixed-dimensional interface, and improve optical gain and lasing behaviour.¹⁰ The mechanism is not enhanced electrical extraction; it is radiative-state preservation through a cleaner and more ordered interfacial landscape. Reduced energetic disorder lowers non-radiative scattering and helps sustain the population inversion or gain-relevant excited-state distribution required for lasing.¹⁰ The device impact is lower threshold and improved optical-gain performance, not stronger PL quenching.¹⁰

Coupling-sensitive interfaces can also create optical functionality beyond simple emission intensity. In chiral perovskite/monolayer MoS₂ heterostructures, spin-selective transfer changes the valley population of MoS₂ and produces room-temperature valley polarization.⁹ The useful device impact is therefore not faster carrier collection, but conversion of interfacial coupling into a polarization-resolved optical response.⁹ This example clarifies why emission and gain interfaces should not be judged by the same criterion as solar-cell contacts. If the interface removes carriers too efficiently into non-radiative or electrically inactive channels, emission, gain, and lasing are suppressed. If it preserves or selectively redistributes radiative excited states, the interface becomes optically functional.^{9,10}

Across these device classes, the same interfacial photophysics produces different consequences because the target output changes. Solar cells reward selective extraction, recombination suppression, and long-term contact stability.^{3,4,6} Photodetectors reveal whether the interface is genuinely extraction-enhancing or merely trap-amplifying through the coupled behaviour of responsivity, speed, and dark current.^{11,37–39} Emission and gain devices reward interfaces that avoid destructive extraction and instead preserve or engineer radiative states.^{9,10} Table 3 summarizes the device-level consequences of representative perovskite–2D interfaces using device-class-specific

quantitative benchmarks. Solar-cell entries include PCE, Voc or Voc loss, FF, operational-stability protocol, device area and certification status where available; photodetector entries include responsivity, detectivity, dark current, response speed, linear dynamic range and persistent-photoconductivity behaviour where reported; and emission/gain entries include gain coefficient, threshold, linewidth and stability-related information where available. Metrics not reported in the original studies are indicated as not reported.

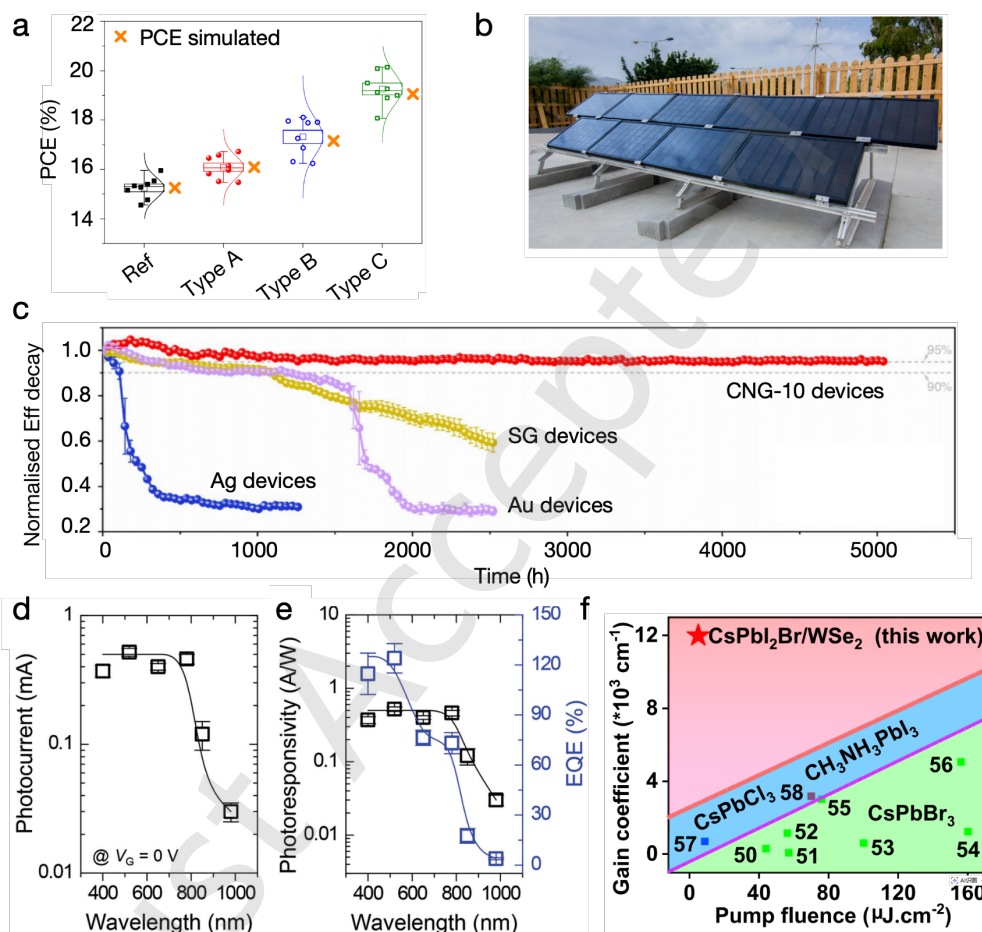


Figure 4 | Device-level impact of interfacial photophysics in perovskite-2D heterostructures. a, MXene-assisted interface engineering improves perovskite solar-cell efficiency by tuning contact energetics and interfacial charge extraction, as reflected by the PCE comparison between reference and MXene-modified device types. Type A, MXene-doped perovskite absorber; Type B, MXene-doped perovskite and TiO₂ layers; Type C, Type B plus an additional MXene interlayer at the mTiO₂/perovskite interface. b, 2D-material-based GRAPE perovskite panels demonstrate translation of interface engineering from small-area cells to outdoor, field-relevant photovoltaic operation. c, Graphene-protected Cu-Ni rear electrodes improve operational stability, with CNG-based devices retaining normalized efficiency far longer than Ag, Au, or single-graphene controls under prolonged operation. d,e, Perovskite-graphene hybrid photodetectors show broadband photocurrent, high photoresponsivity and high EQE, illustrating photogating-amplified detector response enabled by coupling perovskite absorption to a graphene transport channel. f, Facet-

selective CsPbI₂Br/WSe₂ heterostructures exhibit enhanced optical-gain coefficients relative to representative perovskite gain media, showing how coupling-sensitive interfaces can preserve radiative populations and improve gain-relevant device performance. Panel a adapted with permission from Ref. 3 ©2019, Springer Nature. Panel b adapted with permission from Ref. 6 ©2022, Springer Nature. Panel c adapted with permission from Ref. 4 ©2022, Springer Nature. Panel d adapted with permission from Ref. 38 ©2014, Wiley-VCH Verlag GmbH & Co. KGaA. Panel e adapted with permission from Ref. 38 ©2014, Wiley-VCH Verlag GmbH & Co. KGaA. Panel f adapted under the CC BY 4.0 license from Ref. 10 ©2024, The Author(s).

Table 3 | Interface photophysics translated into device-level metrics. Representative systems are compared to show how extraction-dominated, loss-dominated and coupling-sensitive interfacial pathways translate into device-level performance. The key quantitative benchmarks are listed where available; NR denotes metrics not reported in the cited study or not available from the summarized comparison.

Device class	Representative interface / system	Controlled variable	Key quantitative benchmarks	Photophysical origin	Device-level implication	Refs.
Solar cells	Cu–Ni/graphene rear electrode	Bifacial graphene diffusion barrier; Cu–Ni work-function matching	Certified PCE: 20.8% for 0.09 cm ² device and 20.86% for 1.02 cm ² device. Device area: 0.09 and 1.02 cm ² . Stability: retained 97% after 1440 h damp heat at 85 °C/85% RH and 95% after 5000 h MPPT under 1 sun. Voc/Voc loss and FF: NR.	Electrode-facing barrier suppresses oxygen/moisture ingress and parasitic metal/device reactions while preserving contact function.	Improved operational stability without treating graphene merely as a conductive additive.	4
Solar cells	Ti ₃ C ₂ T _x -modified perovskite/TiO ₂ interface	Termination-dependent work-function tuning and perovskite/ETL band alignment	PCE increased by 26% relative to reference cells. Hysteresis was reduced. Numerical Voc/Voc loss, FF, device area, certification status and operational-stability protocol: NR.	Surface terminations shift the work functions of the perovskite absorber and TiO ₂ ETL, reducing the extraction barrier.	Improved charge collection and contact selectivity at the electron-selective interface.	3
Solar cells / tandem modules	Cl-terminated MXene buried interface	Interfacial dipoles; local band bending; n-type buried-interface region	Semitransparent modules achieved >16% PCE on 60 cm ² active area. A 0.2 m ² four-terminal tandem panel reached 19.45% PCE. Stability: retained >95% delivered power after three months outdoors. Voc/Voc loss, FF and certification status: NR.	Dipole-driven local field forms a nanoscale field-effect junction inside the perovskite active layer.	Improved extraction and reduced recombination through electrostatic steering rather than simple planar charge acceptance.	17
Solar cells	WS ₂ /CsPbBr ₃ van der Waals heterostructure	Strain release at ETL/perovskite boundary	PCE increased from 9.27% to 10.65%, corresponding to an	Matched lattice and atomically smooth, dangling-bond-free	Improved device performance through strain-	18

			absolute gain of 1.38 percentage points. Voc/Voc loss, FF, device area, certification status and operational-stability protocol: NR.	WS ₂ interface releases tensile strain in CsPbBr ₃ .	associated defect reduction rather than direct charge-transfer assignment.	
Solar cells	BP-, MoS ₂ -, and graphene-oxide-flake-modified MAPbI ₃ films	Local grain-boundary hole pathway	Device improvement depends on the mobility and location of the 2D flake at grain-boundary edges. A unified PCE/Voc/FF benchmark is NR because the reported metric varies with flake type and device configuration. Stability protocol, device area and certification status: NR.	2D flakes conduct holes from grain boundaries to the HTL, competing with grain-boundary recombination.	Device benefit arises from local carrier routing rather than a uniform planar perovskite/2D heterojunction.	7
Photodetectors	Perovskite-graphene hybrid photodetector	Perovskite absorption coupled to graphene conductance modulation	Responsivity: up to 180 A W ⁻¹ . Effective quantum efficiency: approximately 5 × 10 ⁴ %. Spectral response: broadband. Dark current, detectivity, response speed, linear dynamic range and persistent photoconductivity: NR.	Photogating: trapped photocarriers in perovskite modulate the high-mobility graphene channel.	High responsivity is enabled by gain, but speed must be interpreted with carrier-storage dynamics.	38
Photodetectors	Perovskite-MoS ₂ hybrid photodetector	Perovskite sensitization of MoS ₂ channel; interfacial carrier transfer/gain	Responsivity: 1.94 × 10 ⁶ A W ⁻¹ . Detectivity: 1.29 × 10 ¹² Jones under 520 nm and 4.63 pW illumination. Dark current, response speed, linear dynamic range and persistent photoconductivity: NR.	Hybrid perovskite/TMD interface produces ultrahigh gain through sensitization and interfacial carrier transfer.	Responsivity must be read together with dark current and response speed to distinguish clean extraction from gain/trapping.	39
Photodetectors	MASnI ₃ /MoS ₂ hybrid-dimensional heterojunction transistor	Hybrid-dimensional interface balancing transfer and transit time	Response speed: rise/decay times of 28.7/31.7 ns. Responsivity, detectivity, dark current, linear dynamic range and persistent photoconductivity: NR.	Interface design shortens response time while retaining photoconductive gain.	Shows how device design can reduce the usual responsivity-speed trade-off.	37
Emission / gain / lasing	Facet-selective CsPbI ₂ Br/WSe ₂ heterostructure	Controlled crystal plane; reduced energetic disorder; radiative-state preservation	Urbach energy reduced from 10.4 to 6.5 meV. Surface recombination velocity reduced from 3700 to 1300 cm s ⁻¹ . Optical gain coefficient increased from 8296 to 12,000 cm ⁻¹ . Lasing threshold reduced from 6.36 to 1.98 μJ cm ⁻² . Emission linewidth and operational stability: NR.	Cleaner mixed-dimensional interface preserves gain-relevant radiative population and suppresses disorder-induced loss.	Improves gain/lasing by avoiding destructive extraction and reducing energetic disorder.	10

Table note: NR, not reported in the cited study or not available from the summarized comparison.

5. Outlook

The next stage of perovskite–2D interface research should move from adding new 2D material species to defining programmable photophysical design rules. Cleaner interfaces are the first requirement because extraction, loss, and coupling cannot be separated when the junction contains uncontrolled roughness, trapped residues, or ill-defined contact geometry. Molecularly thin perovskite/graphene contacts show that a clean van der Waals interface can connect barrier reduction, tunnelling, and femtosecond charge transfer in a single physical picture.⁸ Facet-selective perovskite/TMD heterostructures show that controlled interfacial crystal planes reduce energetic disorder and support optical gain.¹⁰ These cases indicate that future interfaces should be designed with defined contact geometry, measured energetics, and reproducible structural registry, rather than treated as chemically modified black boxes.

Measured energetics should become a standard design constraint. Work-function shifts, band offsets, interface dipoles, and local band bending determine whether photoexcited states enter extraction, loss, or radiative/coupled pathways. $\text{Ti}_3\text{C}_2\text{T}_x$ MXenes demonstrate how surface terminations tune the work functions of the perovskite absorber and TiO_2 electron-transport layer, modify band alignment, and improve selective extraction.³ Cl-terminated MXene buried interfaces show that termination-induced dipoles can create local band bending and n-type interfacial character, turning the buried junction into a nanoscale field-effect interface.¹⁷ These examples point to an interface-design rule: energy levels should be measured locally and under relevant stack conditions, rather than inferred from isolated material work functions.

Multimodal spectroscopy is needed to connect those energetic variables to the excited-state pathway. TRPL identifies emissive-state depopulation, TA follows population redistribution, Kelvin-probe or KPFM measurements map local potentials, photocurrent dynamics test carrier collection, and SHG probes symmetry or field-sensitive optical response. The important advance is not simply combining more techniques, but using them on the same interface to decide whether the photoexcited population becomes separated charge, non-radiative loss, or a coupled radiative state. Interfacial recombination imaging and non-radiative loss mapping already show that spatially resolved optical measurements can connect local PL loss to voltage loss.^{21,22} Time-resolved SHG would extend this logic to interfacial polarity and photoinduced symmetry changes, allowing transient fields and coupling-related symmetry responses to be followed on the same timescale as TA and TRPL.^{29,32}

Programmable coupling variables provide the longer-term opportunity. In TMD heterostructures, twist and registry create moiré potentials that localize excitons, split excitonic resonances, and tune hybridized interlayer exciton states.^{36,40} Correlated exciton phases and moiré photonic responses further show that excitonic behaviour can be engineered by geometry rather than by composition alone.^{5,41} Perovskite–2D systems are unlikely to copy TMD moiré physics directly because perovskites are softer, more ionic, and more structurally dynamic. Their opportunity is different: registry, local strain, dielectric screening, and dipolar environment could be used as lower-barrier knobs for controlling interfacial disorder, radiative-state preservation, and field-assisted separation. Dipolar interactions between localized interlayer excitons in van der Waals heterostructures show

that out-of-plane dipoles can tune excitonic interactions; in perovskite–2D interfaces, analogous dipolar environments may become useful for controlling interfacial fields and radiative-state redistribution.⁴²

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None.

Author contribution statement

Dengyang Guo and Bilu Liu conceived the scope and structure of the Review. Di Yang contributed to the device-related sections. Zhiyuan Zhang contributed to the sections on second-harmonic generation and related nonlinear optical characterization. Fu Deng contributed to the sections on interfacial photophysics. Dengyang Guo conducted the remaining literature review and synthesis, prepared the figures and tables, and wrote and integrated the manuscript. All authors reviewed and edited the manuscript and approved the final version.

Data availability

Not applicable.

Declaration of competing interest

Bilu Liu is an editorial board member of this journal, but he was not involved in the peer review or decision-making process for this article. The other contributing authors report no conflicts of interest in this work.

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Use of AI statement

During the preparation of this work, the authors used ChatGPT (OpenAI) to assist with the preparation and refinement of schematic illustrations and with English grammar and language editing. After using this tool, the authors reviewed and edited all relevant content as needed and take full responsibility for the content of the publication.

References

1. Qiu, L. *et al.* Interfacial engineering of halide perovskites and two-dimensional materials. *Chem. Soc. Rev.* **52**, 212–247 (2023).

2. Wang, B. *et al.* The charge carrier dynamics, efficiency and stability of two-dimensional material-based perovskite solar cells. *Chem. Soc. Rev.* **48**, 4854–4891 (2019).
3. Agresti, A. Titanium-carbide MXenes for work function and interface engineering in perovskite solar cells. *Nat. Mater.* **18**, 1228–1234 (2019).
4. Lin, X. In situ growth of graphene on both sides of a Cu–Ni alloy electrode for perovskite solar cells with improved stability. *Nat. Energy* **7**, 520–527 (2022).
5. Du, L. Moiré photonics and optoelectronics. *Science* **379**, eadg0014 (2023).
6. Pescetelli, S. *et al.* Integration of two-dimensional materials-based perovskite solar panels into a stand-alone solar farm. *Nat. Energy* **7**, 597–607 (2022).
7. You, P. *et al.* 2D materials for conducting holes from grain boundaries in perovskite solar cells. *Light Sci. Appl.* **10**, 68 (2021).
8. Leng, K. Electron tunneling at the molecularly thin 2D perovskite and graphene van der Waals interface. *Nat. Commun.* **11**, 5483 (2020).
9. Shrestha, S. *et al.* Room temperature valley polarization via spin selective charge transfer. *Nat. Commun.* **14**, 5234 (2023).
10. Zhang, R. Facet-selective growth of halide perovskite/2D semiconductor van der Waals heterostructures for improved optical gain and lasing. *Nat. Commun.* **15**, 5535 (2024).
11. Wang, Y. *et al.* Solution-Processed MoS₂/Organolead Trihalide Perovskite Photodetectors. *Adv. Mater.* **29**, 1603995 (2017).
12. Qiao, J., Kong, X., Hu, Z.-X., Yang, F. & Ji, W. High-mobility transport anisotropy and linear dichroism in few-layer black phosphorus. *Nat. Commun.* **5**, 4475 (2014).
13. Youngblood, N., Chen, C., Koester, S. J. & Li, M. Waveguide-integrated black phosphorus photodetector with high responsivity and low dark current. *Nat. Photonics* **9**, 247–252 (2015).
14. Tang, G. Solution-Phase Epitaxial Growth of Perovskite Films on 2D Material Flakes for High-Performance Solar Cells. *Adv. Mater.* **31**, 1807689 (2019).
15. You, P., Liu, Z., Tai, Q., Liu, S. & Yan, F. Efficient Semitransparent Perovskite Solar Cells with Graphene Electrodes. *Adv. Mater.* **27**, 3632–3638 (2015).
16. Chen, W. *et al.* Black Phosphorus Quantum Dots for Hole Extraction of Typical Planar Hybrid Perovskite Solar Cells. *J. Phys. Chem. Lett.* **8**, 591–598 (2017).
17. Agresti, A. *et al.* MXene-driven nanoscale field-effect junction for advanced 4-terminal perovskite/silicon tandem solar panels. *Nat. Commun.* **17**, 3394 (2026).
18. Zhou, Y. Interfacial Strain Release from the WS₂/CsPbBr₃ van der Waals Heterostructure for 1.7 V Voltage All-Inorganic Perovskite Solar Cells. *Angew. Chem. Int. Ed.* **59**, 18670–18678 (2020).
19. Hong, X. Ultrafast charge transfer in atomically thin MoS₂/WS₂ heterostructures. *Nat. Nanotechnol.* **9**, 682–686 (2014).

20. Jin, C. Ultrafast dynamics in van der Waals heterostructures. *Nat. Nanotechnol.* **13**, 994–1003 (2018).
21. Stolterfoht, M. *et al.* Visualization and suppression of interfacial recombination for high-efficiency large-area pin perovskite solar cells. *Nat. Energy* **3**, 847–854 (2018).
22. Cacovich, S. *et al.* Imaging and quantifying non-radiative losses at 23% efficient inverted perovskite solar cells interfaces. *Nat. Commun.* **13**, 2868 (2022).
23. Gerhard, M. *et al.* Microscopic insight into non-radiative decay in perovskite semiconductors from temperature-dependent luminescence blinking. *Nat. Commun.* **10**, 1698 (2019).
24. Pan, J. *et al.* Operando dynamics of trapped carriers in perovskite solar cells observed via infrared optical activation spectroscopy. *Nat. Commun.* **14**, 8000 (2023).
25. Guo, D., Caselli, V. M., Hutter, E. M. & Savenije, T. J. Comparing the Calculated Fermi Level Splitting with the Open-Circuit Voltage in Various Perovskite Cells. *ACS Energy Lett.* **4**, 855–860 (2019).
26. Jiang, P. Interlayer exciton formation, relaxation, and transport in TMD van der Waals heterostructures. *Light Sci. Appl.* **10**, 72 (2021).
27. Barré, E. Optical absorption of interlayer excitons in transition-metal dichalcogenide heterostructures. *Science* **376**, 406–410 (2022).
28. Hu, Y. Excitonic Interactions and Mechanism for Ultrafast Interlayer Photoexcited Response in van der Waals Heterostructures. *Phys. Rev. Lett.* **131**, 236904 (2023).
29. Zhang, Z.-Y. *et al.* Thermal expansion mismatch-driven high-pressure growth of 2D non-centrosymmetric MnTeMoO6 with giant nonlinear optical properties. *Matter* **8**, 102247 (2025).
30. Mihalyi-Koch, W. *et al.* Revealing hidden non-centrosymmetry in globally centrosymmetric 2D halide perovskites. *Chem* **10**, 2180–2195 (2024).
31. Zhao, K. Probing the Interfacial Symmetry Using Rotational Second-Harmonic Generation in Oxide Heterostructures. *J. Phys. Chem. C* **123**, 23000–23006 (2019).
32. Seyler, K. L. *et al.* Electrical control of second-harmonic generation in a WSe2 monolayer transistor. *Nat. Nanotechnol.* **10**, 407–411 (2015).
33. Okada, D. & Araoka, F. Electric-Field-Induced Giant Resonant Enhancement of Second Harmonic Generation in Two-Dimensional Hybrid Perovskite. *J. Am. Chem. Soc.* **147**, 14677–14683 (2025).
34. Li, D. *et al.* Polar coupling enabled nonlinear optical filtering at MoS2/ferroelectric heterointerfaces. *Nat. Commun.* **11**, 1422 (2020).
35. Li, D. *et al.* Ferroelectric Domain Control of Nonlinear Light Polarization in MoS2 via PbZr0.2Ti0.8O3 Thin Films and Free-Standing Membranes. *Adv. Mater.* **35**, 2208825 (2023).
36. Jin, C. Observation of moiré excitons in WSe2/WS2 heterostructure superlattices. *Nature* **567**, 76–80 (2019).
37. Zhang, C. Photodetectors Based on MASnI3/MoS2 Hybrid-Dimensional Heterojunction Transistors: Breaking the Responsivity–Speed Trade-Off. *ACS Nano* **18**, 19303–19313 (2024).

38. Lee, Y. High-Performance Perovskite-Graphene Hybrid Photodetector. *Adv. Mater.* **27**, 41–46 (2015).
39. Kang, D.-H. An Ultrahigh-Performance Photodetector based on a Perovskite–Transition-Metal-Dichalcogenide Hybrid Structure. *Adv. Mater.* **28**, 7799–7806 (2016).
40. Alexeev, E. M. Resonantly hybridized excitons in moiré superlattices in van der Waals heterostructures. *Nature* **567**, 81–86 (2019).
41. Xiong, C. Correlated insulator of excitons in WSe₂/WS₂ moiré superlattices. *Science* **380**, 860–863 (2023).
42. Li, H. Dipolar interactions between localized interlayer excitons in van der Waals heterostructures. *Nat. Mater.* **19**, 830–834 (2020).