Filtering the photoluminescence spectra of atomically thin semiconductors with graphene

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Atomically thin semiconductors made from transition metal dichalcogenides (TMDs) are model systems for investigations of strong light–matter interactions and applications in nanophotonics, optoelectronics and valleytronics. However, the photoluminescence spectra of TMD monolayers display a large number of features that are particularly challenging to decipher. On a practical level, monochromatic TMD-based emitters would be beneficial for low-dimensional devices, but this challenge is yet to be resolved. Here, we show that graphene, directly stacked onto TMD monolayers, enables single and narrow-line photoluminescence arising solely from TMD neutral excitons. This filtering effect stems from complete neutralization of the TMD by graphene, combined with selective non-radiative transfer of long-lived excitonic species to graphene. Our approach is applied to four tungsten- and molybdenum-based TMDs and establishes TMD/graphene heterostructures as a unique set of optoelectronic building blocks that are suitable for electroluminescent systems emitting visible and near-infrared photons at near THz rate with linewidths approaching the homogeneous limit.

TMD monolayers (hereafter simply denoted TMDs), such as MoS2, MoSe2, WS2 and WSe2, are direct-bandgap semiconductors featuring short Bohr radii, large exciton binding energy of hundreds of meV (refs. 1,2) and picosecond excitonic radiative lifetimes at low temperature,3, all arising from their strong two-dimensional (2D) Coulomb interactions, reduced dielectric screening and large effective masses3–5. Since the first investigations of light emission from TMDs, it has been clear that their low-temperature spectra are composed of at least two prominent features, stemming from bright neutral excitons (X0) and charged excitons (trions, X*)4–10 endowed with a binding energy of typically 20–40 meV relative to X0. Among the vast family of TMDs, one may distinguish between so-called dark and bright materials9,10. In the case of tungsten-based TMDs, a spin-dark state lies lower than X0. Conversely, in molybdenum-based TMDs, X0 lies below (MoSe2) or very near (MoS2) the spin-dark state, resulting in brighter emission at low temperature. As a result, X0 and X* emission dominate the photoluminescence (PL) spectrum of Mo-based TMDs, whereas the emission spectrum of W-based TMDs display a complex series of lines stemming from X0, biexcitons (XX0)12–15, charged excitonic states (including X*)10,16 and charged biexcitons (XX*)12–15, spin-dark excitons17–19, defect-induced emission20 and exciton–phonon sidebands21,22.

Considerable progress has been made to deterministically observe intrinsic TMD emission features. In particular, encapsulation of TMDs in hexagonal boron nitride (BN) films results in narrower neutral exciton linewidths23,24 approaching the radiative limit23,24, without, however, getting rid of the other emission features mentioned above. Even in electrostatically gated devices tuned near the charge neutrality point, sizeable emission sidebands remain observable at energies close to the X* feature, suggesting residual charge inhomogeneity25,26 or intrinsic contributions from longer-lived exciton–phonon replicas21,22.

The complex emission spectra of TMD stimulate lively scientific debates. Conversely, obtaining atomically thin semiconductors with single, narrow emission lines remains an important challenge in the field. An appealing solution could consist in interfacing a TMD monolayer with graphene. Indeed, the semimetallic character of graphene and its highly symmetric electronic structure27, with its Dirac point lying within the bandgap of Mo- and W-based TMDs, makes it an ideal electron and hole acceptor, through static charge transfer28,29. Unfortunately, at room temperature, the effective X0 lifetime is in the nanosecond range30,31 and interlayer coupling between TMDs and graphene results in significant PL quenching30,32 due to picosecond energy transfer mediated by either charge tunnelling (Dexter-type) or longer-range dipole–dipole interactions ( Förster-type)30,33,34. However, a much more favourable situation may occur at lower temperatures (typically, below 100 K), where the radiative lifetime of X0 drastically shortens30 and becomes of the same order of magnitude as the theoretically estimated energy transfer time35.

In this Article, we demonstrate that W- and Mo-based TMDs coupled to a graphene monolayer exhibit only one single and narrow emission line that is assigned to X0 radiative recombination, indicating complete neutrality. The short-lived X0 states are minimally affected by non-radiative transfer to graphene and subsequent PL quenching, in stark contrast with longer-lived excitonic species, which are massively quenched. Graphene has been recognized as a partner material of choice to improve the optoelectronic response of TMDs, whereas TMDs hold promise to improve spin transport in graphene36,37. Our results now establish TMD/graphene heterostructures as outstanding light-emitting systems, readily interfaced with a quasi-transparent conductive channel.

Bright, single-line emission in TMD/graphene heterostructures.

The lower panel in Fig. 1 shows the PL spectra of van der Waals heterostructures made from monolayers of MoS2, MoSe2, WS2 and WSe2 stacked onto graphene monolayers and encapsulated in BN. These spectra are compared to those of neighbouring BN-capped TMD regions (upper panel). All TMD/graphene spectra display
one single and narrow Lorentzian emission line, with a full-width at half-maximum (FWHM) of typically 5 meV, suggesting minimal dephasing and disorder (see Supplementary Section 1 for the fitting parameters and Supplementary Section 2 for MoSe2/graphene samples with PL linewidths approaching the homogeneous limit). The TMD references also display narrow emission features assigned to X0 (asterisks in Fig. 1, upper panel), but the latter are accompanied by the lower-energy emission lines introduced above3,16,22,23 (see Supplementary Section 3 for their assignments). The sharp PL lines in TMD/graphene are slightly redshifted (by ~10 meV) with respect to X0 in the TMD references. By measuring the temperature-dependent PL spectra and the differential reflectance (DR) spectra of TMD and TMD/graphene, we can unambiguously assign these single lines to X0 (Fig. 2 and Supplementary Section 4). Therefore, we conclude (1) that no X* emission is measured in TMD/graphene and (2) that the X0 redshift in TMD/graphene arises from dielectric screening30,38. Crucially, we note that PL quenching of the X0 line is moderate, systematically of less than one order of magnitude in all samples under study (Fig. 1 and Supplementary Section 1). The difference between PL from TMD and TMD/graphene heterostructures due to fast non-radiative dephasing and disorder (see Supplementary Section 1 for the fitting parameters and Supplementary Section 2 for MoSe2/graphene samples with PL linewidths approaching the homogeneous limit). The TMD references also display narrow emission features assigned to X0 (asterisks in Fig. 1, upper panel), but the latter are accompanied by the lower-energy emission lines introduced above3,16,22,23 (see Supplementary Section 3 for their assignments). The sharp PL lines in TMD/graphene are slightly redshifted (by ~10 meV) with respect to X0 in the TMD references. By measuring the temperature-dependent PL spectra and the differential reflectance (DR) spectra of TMD and TMD/graphene, we can unambiguously assign these single lines to X0 (Fig. 2 and Supplementary Section 4). Therefore, we conclude (1) that no X* emission is measured in TMD/graphene and (2) that the X0 redshift in TMD/graphene arises from dielectric screening30,38. Crucially, we note that PL quenching of the X0 line is moderate, systematically of less than one order of magnitude in all samples under study (Fig. 1 and Supplementary Section 1). The difference between PL from TMD and TMD/graphene heterostructures is particularly striking in the case of W-based TMDs. In these dark materials, hot luminescence from X0 is quite inefficient and lower-lying emission lines dominate the PL spectra, especially in WS2 (Fig. 1 and Supplementary Section 3). As discussed in the following, all these features are much longer-lived than X0 in TMD6,17,20,39 and are thus literally washed out in the emission spectra of TMD/graphene heterostructures due to fast non-radiative transfer to graphene.

**Complete TMD neutralization.** Figure 2 shows PL mapping as well as typical DR and PL spectra recorded on a BN-capped MoSe2/graphene sample deposited on a SiO2 coverslip. The PL spectrum of the BN-capped MoSe2 reference is composed of two lines with similar intensities, shifted by 28 meV. The high- and low-energy PL lines are assigned to X0 and X*, respectively. An X* absorption feature emerges on the DR spectrum, with an amplitude considerably smaller than that of the X0 DR feature (Fig. 2e). In contrast, the BN-capped MoSe2/graphene region of the sample displays only X0 absorption and emission features. As shown in Fig. 2b–d, these observations can be consistently made over the whole (>40 μm²) area of a coupled MoSe2/graphene region. This is an important point, as TMD-based van der Waals heterostructures are known to be spatially inhomogeneous and the observation of ‘trion-free’ spectra might thus be accidental. Furthermore, the absence of X* emission can be exploited as a reliable probe of the coupling between TMD and graphene.

We assign the absence of X* absorption and emission features in the TMD/graphene region to the transfer of all the native dopants in the TMD (either electrons or holes, with a typical density on the order of 10¹⁰–10¹² cm⁻²) to graphene. Such static charge transfer leads to a slight increase in the Fermi level of graphene (typically by less than 100 meV) and to the observation of intrinsic absorption and emission. Alternative scenarios involving residual doping in the TMD but massive quenching of X0 formation and/or radiative recombination can safely be ruled out (Supplementary Section 5.1). TMD neutralization is corroborated by room-temperature Raman scattering30 and PL measurements (Supplementary Section 5.2).

**Photostability and dielectric screening.** To further establish the benefits of coupling TMDs to graphene, we show, in Fig. 3, the PL spectra of the BN-capped MoSe2/graphene introduced in Fig. 2 recorded under continuous-wave (c.w.) photon fluxes (hereafter Φp) at 2.33 eV, spanning more than five orders of magnitude, from 1×10¹⁰ cm⁻²s⁻¹ to 3×10³⁹ cm⁻³s⁻¹, that is, ~1 mW μm⁻². Assuming an absorbance of ~10% at 2.33 eV (ref. 48) and an exciting lifetime of 2 ps in BN-capped MoSe2/graphene (Figs. 4 and 5), we may...
estimate injected hot exciton densities ranging from $2 \times 10^5 \text{cm}^{-2}$ to $6 \times 10^{10} \text{cm}^{-2}$. The $X^0$ PL intensity scales quasi-linearly with $\phi_{ph}$. We note, however, the emergence of two faint PL features. The first is asymmetric, blueshifted by $\sim 120 \text{meV}$ from $X^0$, and its intensity scales linearly with $\phi_{ph}$. We assign this feature to hot luminescence from the neutral excited $X^0_{1s}$ excitons (at $\Delta_{1s-3s} \approx 110 \text{meV}$) and $X^0_{2s}$ excitons (at $\Delta_{2s-3s} \approx 127 \text{meV}$). The exciton binding energy is $E_b = 231 \text{meV}$ in BN-capped MoSe$_2$, with $E_b = 168 \text{meV}$ (ref. 4). Assuming, for simplicity, that $E_b$ scales proportionally to $\Delta_{1s-3s}$, we estimate that the presence of a graphene layer reduces $E_b$ down to $\sim 150 \text{meV}$ (Supplementary Section 6). The second feature is redshifted by $\sim 25 \pm 2 \text{meV}$, that is, a few meV less than the $X^*$ line in MoSe$_2$ (Fig. 2c) and its intensity rises quadratically with $\phi_{ph}$. Therefore, this feature could tentatively be assigned to a biexciton ($XX^*$). However, XX$^*$ have recently been observed in MoSe$_2$ (ref. 41) and WSe$_2$ (refs. 12,15) monolayers and display binding energies of $\lesssim 20 \text{meV}$, significantly lower than the value observed here in a system that undergoes more screening due to the presence of graphene. Thus, the lower-energy feature is tentatively assigned to emission from photocreated trions with a slightly reduced binding energy. As previously reported in other low-dimensional materials (for example, carbon nanotubes$^{42}$), at sufficiently large exciton densities, bimolecular exciton–exciton annihilation (EEA$^{43}$) can create free carriers. Subsequent photon absorption leads to $X^0$ formation.
in Fig. 4a, the TRPL traces in Fig. 4 are simply fit by the convolution of the instrument response function (IRF, grey area). The extracted $\tau_{X0}$ and $\tau_{X^*}$ lifetimes, respectively, are indicated.

Exciton dynamics and exciton transfer time. Figure 4 compares the time-resolved PL (TRPL) of MoSe$_2$ and MoSe$_2$/graphene, recorded at 14 K on another sample deposited on SiO$_2$ and optically excited below the X$^0$ state. As shown in Fig. 4a, the filtering effect evidenced in BN-capped TMD/graphene (Fig. 1) also appears prominently in samples supported by a rougher SiO$_2$ substrate (Supplementary Section 2). A three-level system considering hot excitons, $X_h$, such as finite momentum $s$ within the light cone) and the ground state is shown in the inset of Fig. 4a. As the PL rise time lies below our time resolution for all measurements on this sample, the TRPL traces in Fig. 4 are simply fit by the convolution of the instrument response function (IRF) with an exponential decay. In keeping with previous reports, the X$^0$ exciton lifetime ($\tau_{X0}$) is only $\sim$2.3 ps in bare MoSe$_2$, and can be assigned to the radiative lifetime $\tau_{X0}^{rad}$ (ref. 6), whereas X$^*$ display a much longer lifetime $\tau_{X^*} = 30$ ps. Remarkably, within experimental accuracy, $\tau_{X0}$ has identical values in MoSe$_2$/graphene and in the neighbouring MoSe$_2$ region. This striking result suggests that $\tau_{X0}^{rad}$ is shorter than $\tau_{X0}^{nonrad}$ in MoSe$_2$/graphene and non-radiative transfer of X$^0$ to graphene, is longer than 2 ps. Let us note that $\tau_{X0}^{rad}$ scales as $E_{C-V}^{-2}$ and is thus expected to be longer in MoSe$_2$/graphene than in the nearby MoSe$_2$ reference. Hence, observing similar $\tau_{X0}^{rad}$ in Fig. 4b,c may be coincidental and result from the compensation between the increase of $\tau_{X0}^{rad}$ in MoSe$_2$/graphene and non-radiative transfer of X$^0$ to graphene with an estimated timescale of $\tau_{X0}^{rel} \approx 5$ ps (Supplementary Section 8.1). As a result, the X$^0$ emission yield (defined here as the number of emitted photons divided by the number of cold X$^0$) that is near unity in the bare MoSe$_2$ monolayer remains high, near 50% in MoSe$_2$/graphene (Supplementary Section 8.2).

Although the integrated PL intensity from X$^0$ in MoSe$_2$/graphene on SiO$_2$ is nearly twice that of a close-lying MoSe$_2$ reference (Fig. 4a), the total PL intensity from MoSe$_2$ remains approximately 4.5 times larger than that of MoSe$_2$/graphene. Following generation of X$^0$, one may form X$^0$, X$^*$ and localized excitons (for example, near defects) in bare MoSe$_2$. Localized excitons contribute a low-energy tail from localized (L) states in bare MoSe$_2$ that is massively quenched in MoSe$_2$/graphene. A three-level system is shown in the left inset. As shown in Fig. 4a, the filtering effect evidenced in BN-capped TMD/graphene (Fig. 1) also appears prominently in samples supported by a rougher SiO$_2$ substrate (Supplementary Section 2). A three-level system considering hot excitons, $X_h$, such as finite momentum $s$ within the light cone) and the ground state is shown in the inset of Fig. 4a. As the PL rise time lies below our time resolution for all measurements on this sample, the TRPL traces in Fig. 4 are simply fit by the convolution of the instrument response function (IRF) with an exponential decay. In keeping with previous reports, the X$^0$ exciton lifetime ($\tau_{X0}$) is only $\sim$2.3 ps in bare MoSe$_2$, and can be assigned to the radiative lifetime $\tau_{X0}^{rad}$ (ref. 6), whereas X$^*$ display a much longer lifetime $\tau_{X^*} = 30$ ps. Remarkably, within experimental accuracy, $\tau_{X0}$ has identical values in MoSe$_2$/graphene and in the neighbouring MoSe$_2$ region. This striking result suggests that $\tau_{X0}^{rad}$ is shorter than $\tau_{X0}^{nonrad}$ in MoSe$_2$/graphene and non-radiative transfer of X$^0$ to graphene, is longer than 2 ps. Let us note that $\tau_{X0}^{rad}$ scales as $E_{C-V}^{-2}$ and is thus expected to be longer in MoSe$_2$/graphene than in the nearby MoSe$_2$ reference. Hence, observing similar $\tau_{X0}^{rad}$ in Fig. 4b,c may be coincidental and result from the compensation between the increase of $\tau_{X0}^{rad}$ in MoSe$_2$/graphene and non-radiative transfer of X$^0$ to graphene with an estimated timescale of $\tau_{X0}^{rel} \approx 5$ ps (Supplementary Section 8.1). As a result, the X$^0$ emission yield (defined here as the number of emitted photons divided by the number of cold X$^0$) that is near unity in the bare MoSe$_2$ monolayer remains high, near 50% in MoSe$_2$/graphene (Supplementary Section 8.2).
time of \( \chi^0 \) down to the light cone. This scenario is consistent with the fact that the Förster-type energy transfer time of TMD excitons to graphene is expected to be maximal for zero momentum excitons (here \( \chi^0 \)) and to decrease with exciton momentum\(^{23,24} \). All in all, our results strongly suggest that graphene opens up efficient non-radiative decay pathways that moderately quench \( \chi^0 \) radiative recombination and instead significantly inhibit \( \chi^0 \) formation. Along these lines, we expect that \( \chi^0 \) formation should be more efficiently inhibited in BN-capped samples, where \( \tau_{rel} \) is much longer than in SiO\(_2\)-supported samples and can be experimentally resolved\(^7\).

Efficient hot exciton transfer to graphene. In Fig. 5, we investigate the exciton dynamics of the BN-capped sample discussed in Figs. 1–3, following pulsed optical excitation slightly below the \( \chi^0 \) exciton energy. Importantly, this sample is deposited on a SiO\(_2\) coverslip and therefore immune from the cavity effects discussed in ref.\(^7\). As introduced in Figs. 1–3, \( \chi^0 \) PL is quenched in MoSe\(_2\)/graphene, in contrast with the case of the SiO\(_2\)-supported sample, where \( \chi^0 \) PL enhancement is observed (Fig. 4a). This difference readily suggests that \( \chi^0 \) formation is more efficiently quenched in BN-capped TMD/graphene. The TRPL traces fit by solving the rate equations associated with the three-level system introduced in Fig. 4a, with two characteristic times for \( \chi^0 \) relaxation towards \( \chi^0 \) (or \( \chi^* \)) formation and \( \chi^0 \) (or \( \chi^* \)) decay, respectively. The short and long times correspond to the rise and decay times in the TRPL measurement, respectively\(^7\). Figure 5b,c shows that the PL rise time (\( \sim 1.5 \) ps) is too close to our resolution limit to be accurately resolved in BN-capped MoSe\(_2\)/graphene and is \( 2.4 \) ps in the BN-capped MoSe\(_2\) reference. The PL decay times are \( \sim 2.2 \) ps in MoSe\(_2\)/graphene and \( \sim 5 \) ps in the MoSe\(_2\) reference region, respectively. Interestingly, the \( \sim 5.1 \) ps PL rise time of the long-lived \( \chi^* \) feature (\( \tau_{rel} = 62 \) ps) in BN-capped MoSe\(_2\) closely matches the \( \chi^0 \) decay time. We therefore assign this \( \sim 5 \) ps time to the relaxation of \( \chi^0 \) down to the \( \chi^* \) and \( \chi^0 \) states in BN-capped MoSe\(_2\). Hot exciton transfer results in a shortened \( \chi^0 \) relaxation time (\( \tau_{rel} \approx 5.1 \) ps in BN-capped MoSe\(_2\)/graphene (Fig. 5b)). Finally, the \( \chi^0 \) lifetimes \( \tau_{\chi^0} \) in BN-capped MoSe\(_2\) and MoSe\(_2\)/graphene are assigned to the rise and decay times of their respective TRPL traces and are nearly identical (\( \sim 2.4 \) ps and \( \sim 2.2 \) ps, respectively). With these values, following the same reasoning as for the SiO\(_2\)-supported MoSe\(_2\)/graphene sample discussed in Fig. 4, we can estimate that the \( \chi^0 \) emission yield is also near 50% (Supplementary Section 8.2), demonstrating that non-radiative \( \chi^0 \) transfer has similar efficiencies in BN-capped and SiO\(_2\)-supported MoSe\(_2\)/graphene heterostructures. We conclude that a significant part of \( \chi^0 \) PL quenching is due to efficient non-radiative transfer of \( \chi^0 \) enabled by graphene and that \( \chi^0 \) PL quenching is more efficient in BN-capped MoSe\(_2\)/graphene than SiO\(_2\)/MoSe\(_2\)/graphene due to the slower \( \chi^0 \) relaxation (or equivalently \( \chi^0 \) formation) in BN-capped MoSe\(_2\).

Conclusions and outlook. In closing, we have shown that graphene neutralizes TMD monolayers, leading to the absence of light emission from charged excitonic species. Graphene also enables picosecond non-radiative transfer of TMD excitonic species. Starting from a given initial \( \chi^0 \) density, transfer of \( \chi^0 \) to graphene will reduce the maximum achievable density of \( \chi^0 \) and of other longer-lived neutral excitonic compounds. The radiative recombination of the latter will be strongly quenched by graphene, whereas the former will be minimally affected owing to the picosecond radiative lifetime. As a result, \( \chi^0 \) exclusively contribute to the PL spectra of TMD/graphene heterostructures. The measured \( \chi^0 \) PL intensity is largely determined by the competition between \( \chi^0 \) formation and non-radiative...
X\textsuperscript{0} transfer to graphene rather than by the trade-off between radiative X\textsuperscript{0} recombination and non-radiative X\textsuperscript{0} transfer to graphene.

Graphene is here introduced as narrow-line filter that is naturally tuned to a broad range of emitted photon energies spanning the mid-infrared to the ultraviolet regions. Our 2D design naturally outperforms alternative solutions based on a spectrally narrow interference filter that are considerably bulkier and lack tunability (Supplementary Section 9). Going further, high-speed (up to ~1 THz) photonic and optoelectronic devices with one single bright and nearly lifetime-limited emission line (Supplementary Section 2) can be envisioned using TMD/graphene heterostructures. Such devices may also benefit from the excellent electrical contact, photodetection\textsuperscript{24,25}, electron and spin transport\textsuperscript{6,7}, capabilities offered by TMD/graphene heterostructures. One may also foresee progress in cavity quantum electrodynamics\textsuperscript{8,9}, chiral optics\textsuperscript{10} and opto-valleytronics\textsuperscript{11} by jointly exploiting the simple emission spectra of TMD/graphene heterostructures and their record-high degrees of valley coherence and valley polarization of up to 60\% and 50\%, respectively\textsuperscript{12}.

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Methods
Our model system is a van der Waals heterostructure formed by stacking a monolayer of graphene onto a TMD monolayer using standard methods as in refs. 51,52. In this work, we have investigated MoSe₂-, MoS₂-, WSe₂- and WS₂-based heterostructures encapsulated in hexagonal BN (Figs. 1–3 and 5) or directly deposited on SiO₂ substrates (Fig. 4). All materials were mechanically exfoliated from bulk crystals. Graphene and TMD monolayers were unambiguously identified using room-temperature Raman and PL spectroscopies, respectively. Our samples were investigated at various temperatures (4–300 K) by means of micro-PL and DR spectroscopy using home-built set-ups. Time-resolved PL measurements were performed on MoSe₂-based samples, using a Ti:sapphire oscillator delivering ~2 ps pulses with a repetition rate of 80 MHz and a synchro-scan streak camera with a temporal resolution of ~1.5 ps. All comparisons between results obtained on TMD and on TMD/graphene are based on measurements performed in the same experimental conditions on the same TMD flake partially covered with graphene (for example, Fig. 2).

Data availability
The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
S.B. conceived and led the project, with C.R., D.L. and X.M. supervising the time-resolved PL measurements. E.L. and L.E.P.L. fabricated the samples. E.L., L.E.P.L., C.R., D.L. and S.B. carried out the measurements. E.L., L.E.P.L. and S.B. analysed the data, with input from G.F., C.R., D.L. and X.M. T.T. and K.W. provided high-quality hexagonal BN crystals. S.B. wrote the manuscript, with input from X.M., C.R., E.L. and L.E.P.L.

Competing interests
The authors declare no competing interests.

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Extended Data Fig. 1 | Approaching the homogeneous limit in BN/MoSe₂/graphene. Photoluminescence (PL) spectra of a MoSe₂ monolayer directly deposited on hexagonal boron nitride (BN) (top, blue line), partly covered by a single layer of graphene (1LG middle, orange) and by a bilayer of graphene (2LG, bottom, dark red). The X₀ exciton energy, full-width at half-maximum (FWHM, denoted \( \Gamma_{X₀} \)) and integrated PL intensity (\( I_{X₀} \), in arbitrary units) are indicated. The spectra were recorded at \( T=15 \) K with laser photon energy of 2.33 eV. We observe nearly identical \( \Gamma_{X₀} \), as small as 1.9 meV and 2.0 meV in the 1LG/MoSe₂/BN and 2LG/MoSe₂/BN, respectively. We find that \( I_{X₀} \) is only quenched by a factor 2.2 (resp. 3.0) in 1LG/MoSe₂/BN (resp. 2LG/MoSe₂/BN) with respect to the vacuum/MoSe₂/BN reference. These results demonstrate that minimal X₀ PL quenching and PL linewidths approaching the homogeneous limit can be achieved in MoSe₂/graphene heterostructures without the need for an extra BN top layer. This figure appears as supplementary Fig. 2 in the supplementary information file.
Extended Data Fig. 2 | Trion-free photoluminescence spectra at room temperature. PL spectra of BN-capped TMD/graphene heterostructures compared to those of a nearby BN-capped TMD reference, all recorded in ambient air in the linear regime under continuous-wave laser excitation at 2.33 eV. The $X^0$ PL lines are symmetric in the TMD/graphene heterostructures whereas they exhibit a lower-energy shoulder arising predominantly from trions ($X^*$) in the TMD references. The scaling factors allow estimating the large room temperature PL quenching factors that strongly contrast with the low $X^0$ PL quenching factors observed at cryogenic temperatures (see main text and supplementary Table 1). The $X^0$ lines are slightly redshifted in TMD/graphene, as discussed in the main text and supplementary Section 4. The red lines are multi-Lorentzian fits to the data, with their different components shown with grey dashed lines. Hot luminescence from excited excitonic states (for example, $X^{2s}_{2s}$ and $B$ excitons) is clearly visible in MoS$_2$/graphene and MoSe$_2$/graphene. This figure appears as supplementary Fig. 7 in the supplementary information file.
Extended Data Fig. 3 | Photostability and neutrality under high photon flux at room temperature. (a) Laser power-dependent photoluminescence spectra of a BN-capped WS$_2$/graphene heterostructure compared to a nearby BN-capped WS$_2$ reference, recorded in ambient air using continuous-wave laser excitation at 2.33 eV. The spectra are shown on a semilogarithmic scale and are normalised by the incoming photon flux ($\Phi_{ph}$) and the integration time. $\Phi_{ph}$ is color-coded with a gradient ranging from dark red (low $\Phi_{ph}$ ~ 100 nW/µm$^2$ or equivalently ~ $3 \times 10^{19}$ cm$^{-2}$s$^{-1}$) to yellow (high $\Phi_{ph}$ ~ 1 mW/µm$^2$ or equivalently ~ $3 \times 10^{23}$ cm$^{-2}$s$^{-1}$). PL saturation due to exciton–exciton annihilation is clearly visible in WS$_2$, whereas a quasi linear scaling is observed in WS$_2$/graphene. The PL spectra in WS$_2$ remain quasi symmetric even under high $\Phi_{ph}$, while the PL spectra from the TMD reference exhibit a lower-energy shoulder, assigned to trion ($X^\ast$) emission. The latter grows significantly as $\Phi_{ph}$ increases and ultimately overcomes the $X^0$ line, as shown in (b) on the selected spectra recorded at $\Phi_{ph}$ ~ $2 \times 10^{23}$ cm$^{-2}$s$^{-1}$ and plotted on a linear scale. This figure appears as supplementary Fig. 8 in the supplementary information file.