

Approaching the intrinsic photoluminescence linewidth in transition metal dichalcogenide monolayers

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LETTER

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Abstract

Excitonic states in monolayer transition metal dichalcogenides (TMDCs) have been the subject of extensive recent interest. Their intrinsic properties can, however, be obscured due to the influence of inhomogeneity in the external environment. Here we report methods for fabricating high quality TMDC monolayers with narrow photoluminescence (PL) linewidth approaching the intrinsic limit. We find that encapsulation in hexagonal boron nitride (h-BN) sharply reduces the PL linewidth, and that passivation of the oxide substrate by an alkyl monolayer further decreases the linewidth and also minimizes the charged exciton (trion) peak. The combination of these sample preparation methods results in much reduced spatial variation in the PL emission, with a full-width-at-half-maximum as low as 1.7 meV. Analysis of the PL line shape yields a homogeneous width of 1.43 ± 0.08 meV and inhomogeneous broadening of 1.1 ± 0.3 meV.

Transition metal dichalcogenides (TMDCs) exhibit fascinating physical properties due to their two-dimensional (2D) nature. This is exemplified by their drastically reduced Coulomb screening and pronounced many-body interactions, leading to excitons [1–3] and trions [4] with respective binding energies of hundreds and tens of meV. Additionally, TMDCs show the unique physical properties of valley selectivity [5, 6] and strong light-matter interactions [7], which make these materials intriguing candidates for optical applications ranging from light detection and emission to quantum computing. Developing a full understanding of the photophysics of TMDC excitons and reaching the potential of TMDCs for applications will require producing materials with optical properties dominated by intrinsic factors, such as their radiative lifetime and electron-phonon interactions, rather than by extrinsic factors such as chemical, mechanical, electrostatic, or dielectric disorder. In high quality epitaxial quantum wells, a related low-dimensional system, the intrinsic PL linewidth has been observed [8, 9],

but it is difficult to reach in nano-materials. For example, exciton emission from ultraclean carbon nanotubes can reach the intrinsic lifetime limit only if they are directly grown over an air gap, with little interference from the local chemical and physical environment [10]. In the case of TMDC monolayers, the extreme sensitivity of atomically thin materials to environmental effects and external disorder [11] has hindered observation of photophysical properties in the intrinsic regime.

The photoluminescence (PL) linewidth observed for exciton emission is a crucial indicator of material quality, and reflects intrinsic contributions from the radiative lifetime and dephasing from exciton–phonon interactions, as well as extrinsic inhomogeneous broadening effects from factors such as defects and substrate-induced disorder. For TMDC monolayers supported on SiO₂ substrates, the PL linewidth is typically exceeds 10 meV at low temperatures. Recent studies employing four-wave mixing techniques indicate that this linewidth is dominated by inhomogeneous broaden-

ing, with an underlying homogeneous contribution of about 2 meV [12–16]. Likewise, the measured exciton emission time has exhibited a large variation associated with significant nonradiative decay channels [12, 17–19]. These studies provide strong motivation to identify methods of sample preparation through which the intrinsic PL linewidth can be reached. An additional important parameter indicative of sample quality in TMDCs is the ratio between the neutral exciton and trion intensity, which has been seen to be strongly dependent on carrier density: the neutral exciton dominates at low density and the trion at high density [20]. It is not presently clear to what degree the carrier density in SiO₂-supported TMDCs results from intrinsic doping due to vacancies and impurities versus from extrinsic doping (from substrate charge traps).

In this work we have systematically investigated the effects of h-BN encapsulation and SiO₂ surface passivation on important features—linewidth, trion/neutral exciton ratio (indicative of doping), and lateral spatial inhomogeneity—of the PL spectrum of a model TMDC, monolayer MoSe₂. Hexagonal boron nitride (h-BN) has proven to be a near-ideal substrate for 2D materials such as graphene and TMDCs. By isolating the materials from the charge disorder at the surface of SiO₂, h-BN encapsulation has been demonstrated to reduce doping and charge inhomogeneity, thus dramatically increasing electron mobility [21–23]. As a consequence, h-BN encapsulation has become standard practice in the fabrication of high-quality 2D devices for electronic applications. Substrate surface passivation by self-assembled monolayers has also been demonstrated as an effective means to increase electron mobility and realize previously elusive *n*-type doping in organic semiconductor FET devices [24]. This was achieved by SiO₂ surface passivation with an organic dielectric monolayer that eliminated hydroxyl groups, and thus electron traps, from the SiO₂ surface. Despite the benefits of surface passivation and h-BN encapsulation in electronic applications, these practices have not yet been widely adopted or investigated in optical studies of TMDCs.

In the present work, we fabricated and investigated four types of samples: (i) monolayer MoSe₂ on an untreated SiO₂/Si substrate (M/SiO₂); (ii) monolayer MoSe₂ on a passivated SiO₂/Si substrate (M/P-SiO₂); (iii) monolayer MoSe₂ doubly encapsulated with h-BN on an untreated SiO₂/Si substrate (BMB/SiO₂); and (iv) monolayer MoSe₂ doubly encapsulated with h-BN on a passivated SiO₂/Si substrate (BMB/P-SiO₂). We mechanically exfoliated MoSe₂ monolayers from a commercially obtained bulk crystal (HQ Graphene) onto an SiO₂/Si substrate (with a 285 nm thick SiO₂ layer) using a combination of oxygen plasma treatment and heating that has been shown [25] to produce larger monolayer flakes. We also mechanically exfoliated high-quality h-BN layers (without heating) and characterized them with atomic force microscopy to identify flakes 20–30 nm thick with clean and ultra-flat areas 20–25 μ m

in size, an important requirement for minimizing sample disorder. Encapsulated h-BN/MoSe₂/h-BN samples were prepared using a dry transfer method with a PC/PDMS lens (PC-poly(bisphenol A carbonate) & PDMS—poly(dimethyl siloxane)) for sequential pickup of the flakes [22]. The pickup process was carried out slowly, with heating/cooling at a rate of ~ 0.5 °C min^{−1} and a vertical translation rate of ~ 0.25 μ m s^{−1} to minimize cracks and trapped bubbles at the h-BN/MoSe₂ interface that could lead to anomalous signatures in our TMDC's PL features. Passivated SiO₂/Si substrates were prepared as follows: the substrates were cleaned by rinsing with CH₂Cl₂, dried, and then oxidized in 3:1 conc. H₂SO₄: 30% H₂O₂ for 2 h at 100 °C, followed by thorough rinsing with DI H₂O. (Caution: the so-called piranha solution is a strong oxidant and should be handled with care.) The self-assembled monolayer (SAM) coating on cleaned SiO₂ surface was obtained from MicroSurfaces, Inc. (<http://microsurfacesinc.com>); the hydrophobic surface is characterized by a water contact angle of $\sim 100^\circ$.

We carried out PL measurements at a temperature of 3.8 K using a closed-cycle He cryostat system with piezo-scanner for mapping (Attocube Attodry 1100). For the PL measurements, the samples were illuminated at a wavelength of 532 nm at low power with a cw diode laser (~ 5 nW for PL intensity mapping and typically < 4 μ W for the PL spectra). The spectra were captured on a liquid nitrogen cooled silicon CCD camera after being dispersed in a grating spectrometer with 70 μ eV resolution at the relevant wavelengths. For intensity mapping, the spectral emission was coupled into a multimode fiber and sent to a silicon single photon counting avalanche photodiode.

In figure 1 we present typical PL spectra for the four different types of sample. Each spectrum shows two peaks, corresponding to the neutral exciton (the higher energy peak) and trion (the lower energy peak) [20]. For the bare monolayer on an untreated substrate (M/SiO₂), the peaks have a full-width at half maximum (FWHM) linewidth of ~ 9 meV. The trion peak shows $\sim 50\%$ greater intensity than the exciton, indicating substantial static doping (figure 1(a)). In addition, a low-energy ‘tail’ extends more than 50 meV below the trion line. h-BN encapsulation (BMB/SiO₂, figure 1(b)) narrows both exciton and trion peaks substantially (to 3 meV), with a clear decrease in the low-energy tail. SiO₂ surface passivation (M/P-SiO₂, figure 1(c)) narrows the peak width to a lesser degree (to 6 meV), but dramatically reduces the trion intensity. Finally, combining both treatments (BMB/P-SiO₂, figure 1(d)) results in the lowest trion intensity and narrowest emission line (2 meV). This linewidth is largely unchanged with increasing temperature up to ~ 100 K, above which it smoothly increases to reach a value of 30 meV at room temperature (figure S1 (stacks.iop.org/TDM/4/031011/mmedia)). We also observe that the exciton emission energy from BNB/P-SiO₂ in figure 1(d) is red-shifted by about 10 meV compared to other samples, likely due

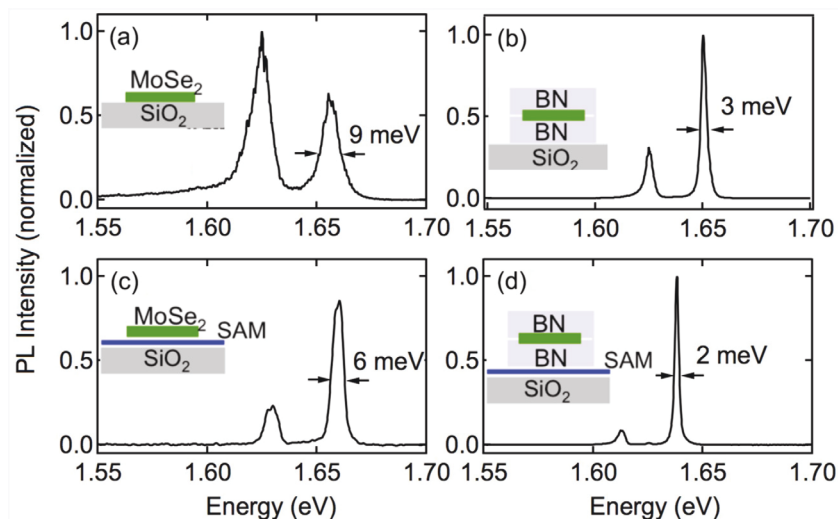


Figure 1. Representative PL spectra of MoSe₂ monolayer at 4K: (a) monolayer on an untreated SiO₂ substrate (M/SiO₂); (b) h-BN-encapsulated monolayer on an untreated SiO₂ substrate (BMB/SiO₂); (c) monolayer on a passivated SiO₂ (M/SiO₂) substrate; (d) h-BN-encapsulated monolayer on a passivated SiO₂ substrate (BMB/SiO₂).

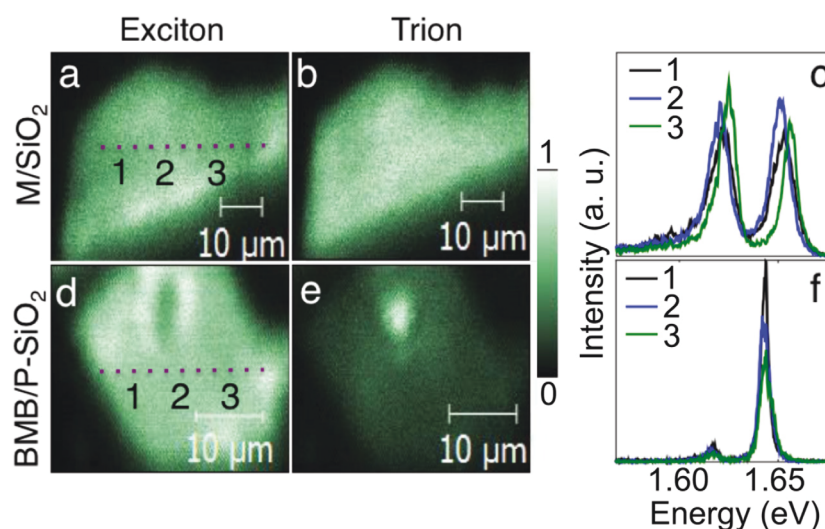
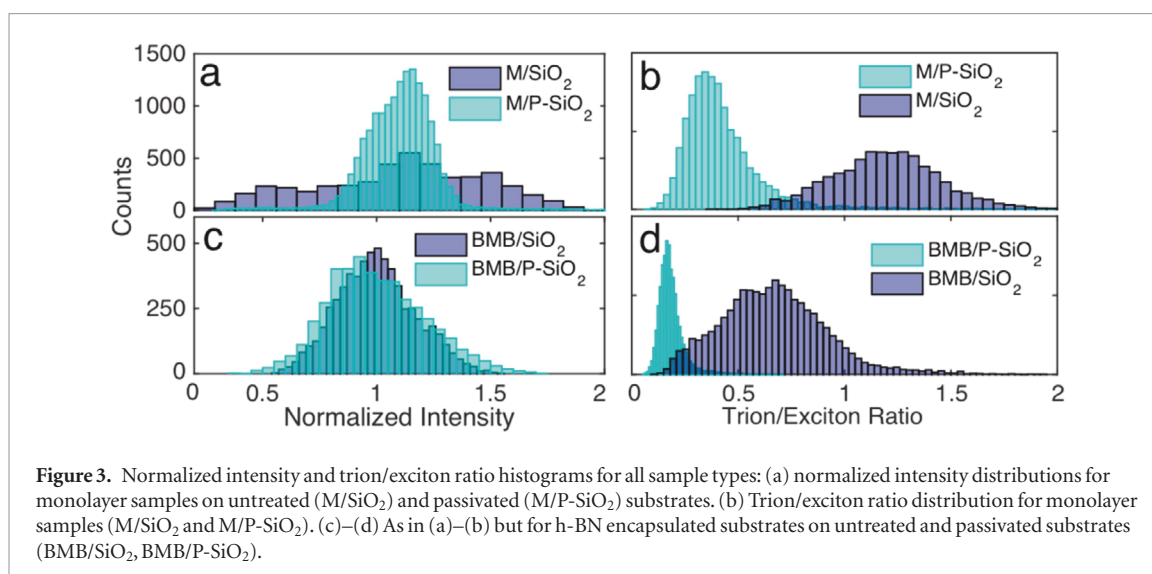


Figure 2. Spatial intensity mapping of exciton and trion emission and spectral linescans for a MoSe₂ monolayer. (a) Exciton spatial map for a monolayer on an untreated SiO₂ substrate (M/SiO₂). (b) Trion spatial map for a monolayer on an untreated SiO₂ substrate (M/SiO₂). (c) PL spectra at the positions indicated in (a) for the M/SiO₂ sample. (d) Exciton spatial map of an h-BN-encapsulated monolayer on a passivated SiO₂ substrate (BMB/P-SiO₂). (e) Trion spatial map of an h-BN-encapsulated monolayer on a passivated SiO₂ substrate (BMB/P-SiO₂). (f) PL spectra at positions indicated in (d) for the BMB/P-SiO₂ sample.

to a reduction in strain [26–28] of the BMB stack on the hydrophobic treated SiO₂ substrate. These findings suggest that the dominant effect of h-BN-encapsulation is to reduce the charge disorder by spatially separating the TMDC layer from the charged SiO₂ surface, giving rise to a significantly narrower linewidth. Similarly, passivation of the SiO₂ improves the emission characteristics predominantly by reducing static charging. The combination of h-BN-encapsulation and SiO₂ surface passivation yields the narrowest PL linewidth for the WSe₂ monolayer, with an emission width approaching the intrinsic limit [12–16], and also drastically reduces the remaining trion intensity (figure 1(d)) which is more than one order of magnitude lower than that on the bare

SiO₂ surface (figure 1(a)) may be attributed to intrinsic doping from structural defects in the WSe₂ monolayer.

In order to study the lateral spatial variation of these features across the sample, we recorded hyperspectral images of the exciton and trion features. This was accomplished by sending the PL through narrow bandpass filters (FWHM = 10 nm) centered at the respective peaks at 750 nm (1.653 eV) and 765 nm (1.620 eV) before reaching the detector. The FWHM of these filters was 10 nm. In figures 2(a) and (b) we present normalized exciton and trion spatial intensity maps for a representative M/SiO₂ sample. We observe comparable exciton/trion intensities over the entire sample, with substantial intensity variation on the scale of a few μm . For a BMB/P-SiO₂ sample, we see (figures 2(d) and (e))



reduced intensity variation and trion intensity, except for a prominent bright trion spot (with a corresponding dark spot in the exciton intensity) that results from a gold alignment marker on the surface. Figures 2(c) and (f) show spectra taken at the three indicated locations on each sample. For the M/SiO₂ sample, we observe fluctuations of about 5 meV in the position of the emission peaks, whereas the BMB/P-SiO₂ sample exhibits no measurable variation in peak position. We note that this fluctuation is significantly smaller than the FWHM of the filters (10 nm–22 meV) used to map the intensity of the exciton and trion peaks. In the following, we present more quantitative data on the spatial variation in the emission characteristics of the different samples.

In figure 3, we display histograms of total PL intensity ((a), (c)) and exciton/trion ratio ((b), (d)) from all the pixels in the PL images in the wrinkle/tear-free locations away from the edges. Both SiO₂ surface passivation and encapsulation dramatically reduce the intensity variation, with the standard deviation dropping by from $41 \pm 10\%$ on M/SiO₂ to $19 \pm 2\%$ on M/P-SiO₂, BMB/SiO₂, or BMB/P-SiO₂. A particularly significant change is in the trion/exciton ratio with the passivation of the SiO₂ surface. This 1.2 ± 0.3 for M/SiO₂ and 0.7 ± 0.3 for BMB/SiO₂ to 0.4 ± 0.2 for M/P-SiO₂ and 0.18 ± 0.06 for BMB/P-SiO₂.

These results establish that h-BN-encapsulation and SiO₂ surface passivation both reduce broadening and the trion/exciton ratio in PL spectra. The best case scenario is obtained when we combine the two approaches and resulting exciton PL linewidth (2 meV) comparable to the homogeneous limit obtained in four-wave mixing experiments [12–16]. In the following, we carry out a quantitative analysis of the exciton PL peaks for BMB/P-SiO₂ samples.

Figure 4(a) shows representative PL spectra in the region of the exciton peak from 10 random locations on a BMB/P-SiO₂ sample. We plot the spectra on a logarithmic intensity scale to reveal weak features in the wings of the spectra. Firstly, we observe variation in the peak exciton energy from different positions.

We find that six of the ten spectra are characterized by excitonic peak positions of $E_{\text{ex}} = 1.6391 \pm 0.0001$ eV, four are red-shifted by as much as 0.003 eV (e.g. the red spectrum in figure 4(a)). Thus we conclude that, while the majority of locations on the BMB/P-SiO₂ sample show homogeneity in PL emission energy, there are also locations with red-shifts in the excitonic transition; the latter likely result from variations local strain [26–28]. Secondly, we note the asymmetry in the peak shape for intensities over one-order-of-magnitude lower than that of the main exciton transition. The intensity is enhanced on the low energy side, which may reflect phonon side bands of the PL emission. The energy scale of this enhancement is consistent with the acoustic phonon energies in MoSe₂ [29]. Thirdly, there is an additional high-energy shoulder, with an intensity reduced more than one order of magnitude lower and peak positions varying from spectrum to spectrum. The origin of this high-energy shoulder is not known, but the variable position suggests that its origin is extrinsic. For instance, a local ‘bubble’ with trapped contaminants such as aromatic hydrocarbons could increase the local dielectric constant and blue-shift the exciton energy [30, 31].

Figures 4(b) and (c) show two representative PL spectrum from the group of six spectra with $E_{\text{ex}} = 1.6391 \pm 0.0001$ eV in figure 4(a) presented on a linear scale. Figures 4(b) is a spectrum with a clearly visible high-energy shoulder, while this feature is nearly invisible in the spectrum in figure 4(c). The intrinsic line-shape from exciton emission is commonly assumed to be Lorentzian, but this may be modified in a TMDC monolayer due to exciton–phonon scattering at non-zero temperatures, exciton–charge scattering in the presence of finite doping, and exciton relaxation to lower energy states such as spin-forbidden dark excitons. While these issues deserve further quantitative studies, here adopt a first-order approximation and assume that each narrow PL peak comes from an intrinsic Lorentian shape broadened by a Gaussian distribution due to inhomogeneity. We fit the main exci-

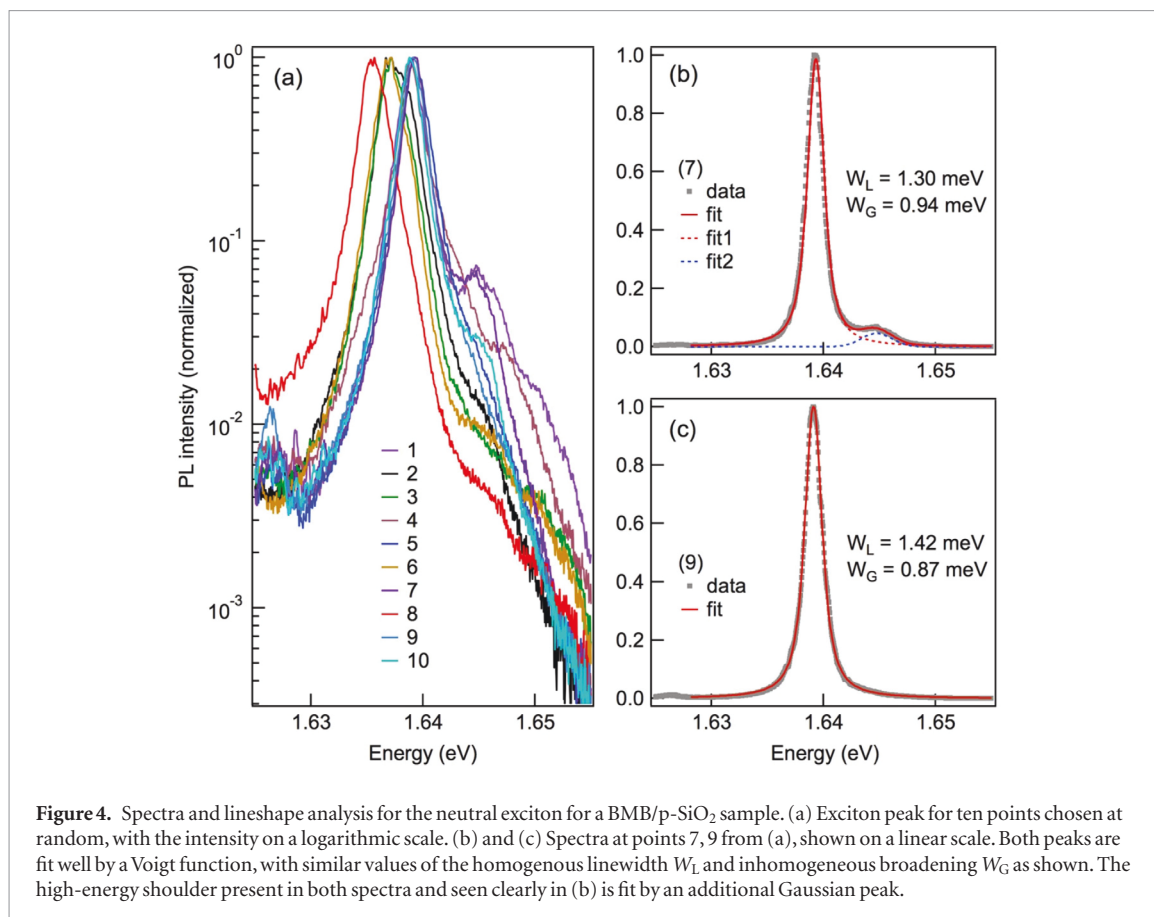


Figure 4. Spectra and lineshape analysis for the neutral exciton for a BMB/p-SiO₂ sample. (a) Exciton peak for ten points chosen at random, with the intensity on a logarithmic scale. (b) and (c) Spectra at points 7, 9 from (a), shown on a linear scale. Both peaks are fit well by a Voigt function, with similar values of the homogeneous linewidth W_L and inhomogeneous broadening W_G as shown. The high-energy shoulder present in both spectra and seen clearly in (b) is fit by an additional Gaussian peak.

Table 1. Summary of homogeneous and inhomogeneous broadening in all sample types.

Sample type	Total emission linewidth (FWHM, meV)	Lorentzian width (meV)	Gaussian width (meV)
M/SiO ₂	9.8 ± 2.8	3.7 ± 0.7	6.5 ± 0.9
M/P-SiO ₂	5.7 ± 1.3	2.9 ± 0.5	3.2 ± 1.3
BMB/SiO ₂	3.5 ± 1.0	1.7 ± 0.5	2.5 ± 0.9
BMB/P-SiO ₂	2.0 ± 0.2	1.43 ± 0.08	1.1 ± 0.3

ton peak with a Voigt function, i.e. a convolution of a Lorentzian and a Gaussian, to represent the influence of homogeneous and inhomogeneous broadening. We note that neither of these functions alone can accurately fit the entire peak, as shown in the supplementary data (figure S2). We add a Gaussian feature (blue-dotted spectrum) to the fit to represent the secondary peak. The fits yield FWHMs of the Lorentzian (W_L) and the Gaussian (W_G) of the main exciton peaks, as shown in figures 4(b) and (c). From fits to all ten spectra, we obtain $W_L = 1.43 \pm 0.08$ meV and $W_G = 1.1 \pm 0.3$ meV. The average value of the total FWHM is 2.0 ± 0.2 meV, with the narrowest line displaying FWHM = 1.7 ± 0.1 meV.

We summarize in table 1 the measured FWHM, along with the Lorentzian (W_L) and Gaussian (W_G) width from the fits for samples prepared according to the different protocols. For the most favorable case of h-BN encapsulated samples on the passivated substrates (BMB/P-SiO₂), we consistently find an inhomogeneous linewidth (W_G) smaller than the homogeneous width (W_L), indicating that the PL spectrum from the

combined h-BN-encapsulation and SiO₂ surface passivation method is approaching the intrinsic limit.

In summary, we show that the use of h-BN encapsulation and substrate surface passivation drastically reduces disorder in WSe₂ monolayers, leading to an 80% reduction in the PL linewidth from that of monolayer on the untreated SiO₂ layer. The PL linewidth for monolayer TMDC on the commonly used SiO₂ surface is dominated by heterogeneity and disorder. We also show that SiO₂ surface passivation and h-BN-encapsulation effectively reduce electron doping of the WSe₂ monolayer by more than one order of magnitude based on the decrease in the strength of trion emission. By strongly reducing local disorder in the substrate, we demonstrate a very significant reduction in the inhomogeneous linewidth. We have achieved an emission linewidth 2.0 ± 0.2 meV, which is predominantly determined by the homogeneous linewidth of 1.43 ± 0.08 meV according to our line shape analysis. During the writing of this manuscript, we became aware of work reporting a similar PL linewidth reduction for MoS₂ monolayers encapsulated by hBN [32].

The realization of samples with such spectrally narrow spectral features should assist in emerging applications of these materials in optoelectronics and valleytronics, as well as in fundamental studies of exciton-charge, exciton-phonon, and exciton-exciton interactions, as well as in interlayer charge transfer exciton formation at TMDC heterojunctions [33]. As one example, narrow linewidth will facilitate accurate measurement of the exciton dispersion, which is predicted to be of order 10 meV [22].

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