

Ultralocalized Optoelectronic Properties of Nanobubbles in 2D Semiconductors

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Cite This: *Nano Lett.* 2022, 22, 7401–7407



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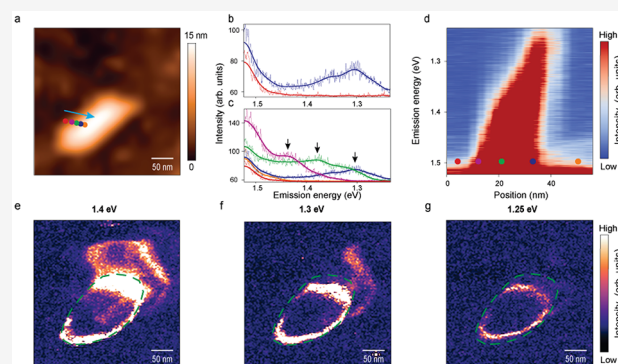
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Supporting Information

ABSTRACT: The optical properties of transition-metal dichalcogenides have previously been modified at the nanoscale by using mechanical and electrical nanostructuring. However, a clear experimental picture relating the local electronic structure with emission properties in such structures has so far been lacking. Here, we use a combination of scanning tunneling microscopy (STM) and near-field photoluminescence (nano-PL) to probe the electronic and optical properties of single nanobubbles in bilayer heterostructures of WSe₂ on MoSe₂. We show from tunneling spectroscopy that there are electronic states deeply localized in the gap at the edge of such bubbles, which are independent of the presence of chemical defects in the layers. We also show a significant change in the local band gap on the bubble, with a continuous evolution to the edge of the bubble over a length scale of ~ 20 nm. Nano-PL measurements observe a continuous redshift of the interlayer exciton on entering the bubble, in agreement with the band-to-band transitions measured by STM. We use self-consistent Schrödinger-Poisson simulations to capture the essence of the experimental results and find that strong doping in the bubble region is a key ingredient to achieving the observed localized states, together with mechanical strain.

KEYWORDS: nanobubbles, 2D semiconductors, ultralocalized, optoelectronic properties, band gap



Two-dimensional (2D) transition-metal dichalcogenides (TMDs) have been investigated as a possible route toward producing well-controlled optical emitters, critical components in quantum technologies and photonics.^{1–12} The band gap of monolayer TMDs is uniquely sensitive to doping,^{13–17} dielectric environment,¹⁸ and mechanical strain,^{8,11,19–21} thereby providing an ideal platform to create quantum confinement by local mechanical and electrical fields. Indeed, previous research has investigated moiré patterns,^{22–26} wrinkles,^{2,4,8} bubbles,^{1,27} and lithographically fabricated dielectric arrays as effective methods²⁸ to create localized optical emitters. In spite of these notable achievements, a basic band diagram picture across such localized structures remains elusive. For example, recent theoretical calculations^{28–30} have predicted large band gap changes at the edges of wrinkles and nanobubbles. However, such predictions have not been experimentally verified, and it remains unclear what the major contributors are to achieving localized optical emitters in TMD materials.

In order to address these questions, high spatial resolution spectroscopic measurements across local perturbations in TMDs are necessary. Our approach is to use the local imaging and spectroscopic capabilities of scanning tunneling microscopy (STM) to measure the local band diagram across

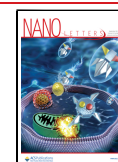
nanobubbles and directly correlate these measurements with near-field optical measurements with 10 nm spatial resolution.

Our STM measurements are conducted on nearly aligned (61.7°) WSe₂/MoSe₂ hetero-bilayers that were stacked on graphite/hBN as a conducting electrode for STM measurements (see Figure 1a for a schematic). The relative orientation between two TMD monolayers was determined by second harmonic generation (SHG) (see Supporting Information Figure S1 and the accompanying discussion). The details of sample fabrication are presented in Methods. During the stacking process, nanobubbles are commonly formed between layers. Figure 1b shows a large-scale atomic force microscopic (AFM) image of such nanobubbles. The nanobubbles range in size from 20 to 400 nm and in height from 2 to 50 nm. STM topographic measurements on one such nanobubble are shown in Figure 1c. Figure 1d shows a height profile of the nanobubble boundary corresponding to the dashed arrow in

Received: June 5, 2022

Revised: September 14, 2022

Published: September 19, 2022



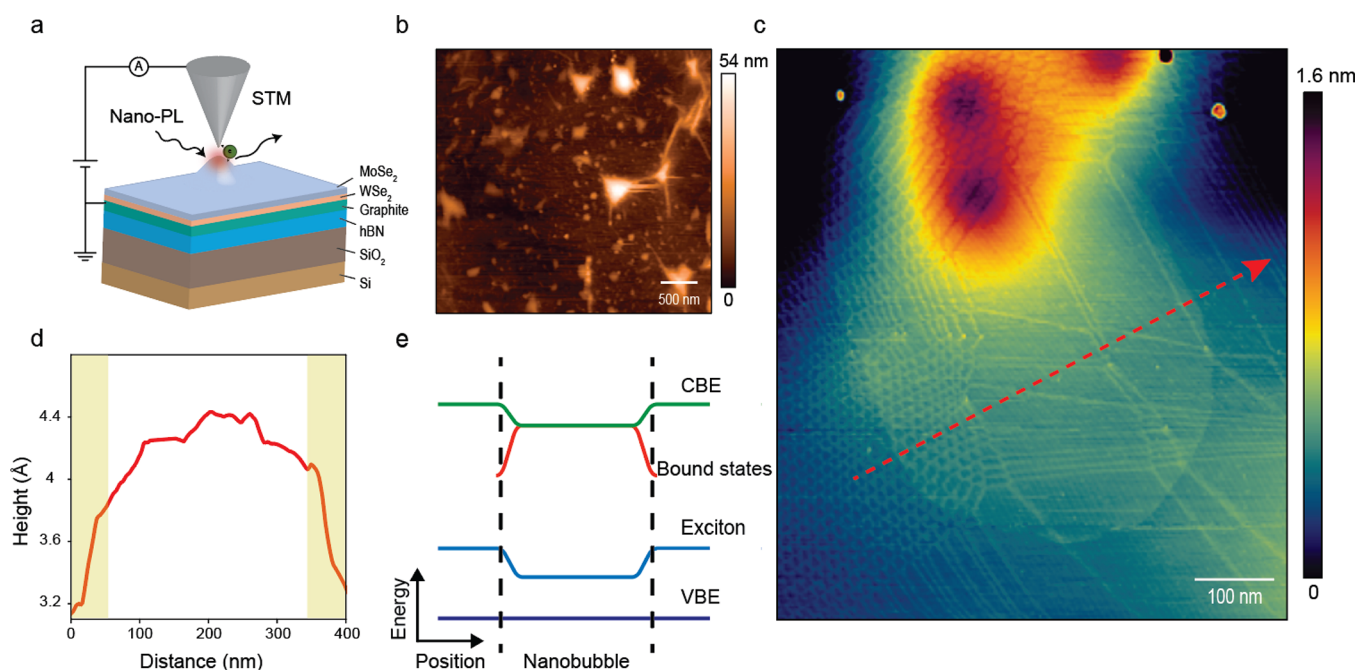


Figure 1. Nanobubbles in twisted WSe₂/MoSe₂ hetero-bilayers. (a) Schematic of the experimental setup. (b) 3.8 μm AFM image of a WSe₂/MoSe₂ hetero-bilayer on gold substrate used in nano-PL measurements showing several nanobubbles. (c) STM topographic image ($V_b = -1.8$ V, $I = -50$ pA) of a single nanobubble. The continuous moiré pattern across the nanobubble shows that contact between the two TMD layers is maintained throughout. (d) STM height profile across the dashed arrow in (c) showing an apparent step size of 1 Å at the nanobubble edge highlighted in yellow. (e) Schematic band diagram of the CB, the bound states, the VB, and interlayer exciton energy as a function of position deduced from STM and nano-PL measurements.

Figure 1c. In order to obtain the STM topography, the tunneling current remains constant at a particular voltage bias, through a feedback loop; however, the tunneling current depends on the integrated local density of states between the sample Fermi level and the bias voltage. This dependency of current on electronic structure affects the topographic data. Thus, both electronic structure and actual topographic height influence the apparent topographic height in STM, and the apparent heights are not directly translatable into real vertical displacements. This consideration is not relevant to AFM measurements; however, AFM measurements are dictated by tip–sample force interactions, which can strongly influence the apparent height in AFM topographic scans. However, it is clearly evident from the line profile across the boundary shown in **Figure 1d** that the edge of the bubble features a sharp step. Such a sharp step indicates that there is localized strain at the edge of the bubble. Finally, a moiré pattern is clearly visible both on and off the bubble. Its presence in all regions shows that the WSe₂ and MoSe₂ layers are in good contact throughout, and the bubble is under the hetero-bilayer. As a result of the device fabrication process under ambient conditions, compounds in air such as water molecules often are trapped between layers. While the chemical identity of the material in the bubble is unknown, our data below, which show a highly doped region on the nanobubble, indicate the presence of polarized (or ionized) molecules trapped in the nanobubble during the fabrication. Such polarized molecules induce doping and electric field to the layers above and modify the electronic structure.

To characterize the electronic properties in the vicinity of the nanobubble, we perform STM dI/dV spectroscopy measurements at various points on and off the bubble (see **Figure 2a**). Shown in **Figure 2b,c** are a sequence of such

measurements taken across the edge of the bubble, color coded according to the markers on **Figure 2a**. **Figure 2b** shows the typical spectra well inside (blue) and outside (red) the nanobubble area. It is seen that the conduction band (CB) edge shows a substantial shift from the outer edge of the bubble into the interior, while the valence band (VB) is nearly unchanged. This indicates a substantial reduction in the band gap on the bubble as well as a large electron doping.

Shown in **Figure 2c** is a sequence of spectra taken in the transition region across the bubble edge, with a focus on the CB edge where the most dramatic changes are observed. It is seen that, at the edge of the bubble (red curve), electronic states exist that are deeply bound in the semiconductor gap. As we transition from outside to inside the bubble (green curve), these states move toward the CB edge, while at the same time the CB edge moves toward the Fermi level. The entire evolution is shown as a heat map of dI/dV in **Figure 2d** as a function of position along the arrow overlaid on **Figure 2a**. The dashed lines overlaid on **Figure 2e** indicate the evolution of the localized states and the conduction band edge upon moving from the outside to the inside of the bubble.

In order to confirm that the observations above are representative of the entire bubble and not specific to a particular region, we performed spectroscopic imaging experiments across the bubble interface at various energies, a subset of which is shown in **Figure 2e–g**. At the valence band edge (**Figure 2e**), the map does not distinguish between the interior and exterior of the bubble, showing the uniformity of the valence band edge across the bubble. **Figure 2f,g**, taken within the semiconducting gap, shows the presence of the localized states clearly and their evolution toward the interior of the bubble. At large positive energies (not shown), these localized states merge into the conduction band, and a clear contrast is

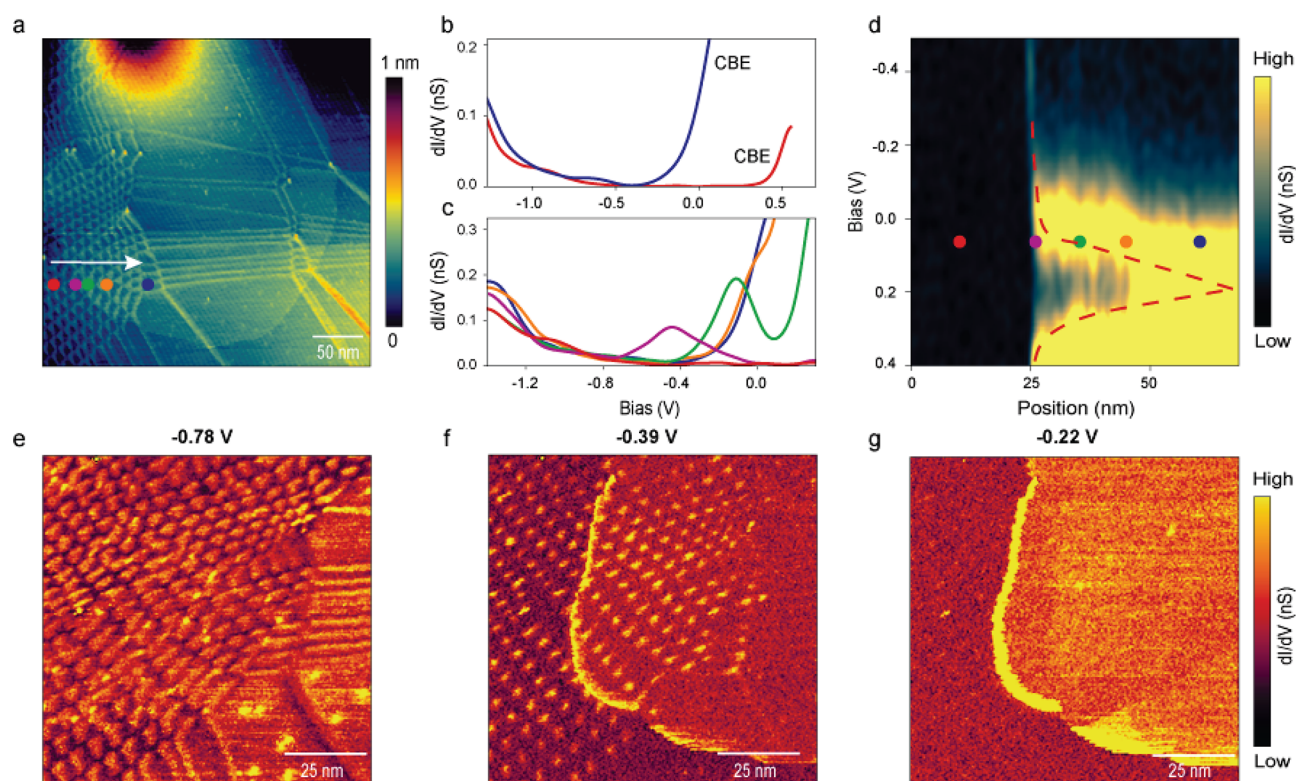


Figure 2. STM spectroscopic properties of nanobubbles. (a) STM topography indicating locations where the point spectra shown in (b, c) were taken. (b) Point spectra taken inside (blue) and outside (red) the bubble show a pronounced shift of the CB edge inside the bubble while the VB is unaffected. (c) Evolution of dI/dV point spectra from the outside to the inside of the nanobubble corresponding to the markers in (a). At the edge of the bubble, deeply localized states are seen within the semiconductor gap. (d) Heat map of position-dependent dI/dV spectra at the CB edge across the bubble. The dashed lines are guides to the eye that show the evolution of the bound states and the CB edge. (e–g) Spectroscopic maps across the nanobubble edge at energies of -0.78 V (VB edge), -0.39 V, and -0.22 V (within the band gap). The set points are $V_b = -1.8$ V and $I = -200$ pA. Bound states are clearly observed at the nanobubble edge at -0.39 and -0.22 V.

seen between the interior and exterior of the bubble. These maps confirm the basic picture provided by point spectra—the bubble region is characterized by localized states at its edge and a shifted conduction band edge throughout. We present larger maps across the entire bubble in the [Supporting Information](#), to rule out moiré effects on the observed phenomena. Finally, to rule out possible damage to the bubble during spectroscopic maps, we measure the topographic heights before and after spectroscopy is performed.

We next proceed to compare our measurements of the single-particle spectroscopic properties of individual bubbles with their optical emission. To do this, we employed hyperspectral nanoscale photoluminescence measurements to map the changes in exciton energy across the bubble. We have previously applied this technique to the imaging of localized exciton states in transition-metal dichalcogenide monolayers;²⁷ however, the spatial resolution was limited due to the finite radius of curvature of the probe. To push the nano-optical resolution to scales on the order of the localized state, we prepared hetero-nanobubbles on template-stripped gold (TS-Au), which allows for optical resolutions on the order of the gap formed between the nano-optic probe and the substrate.³¹ In addition, the quenching of photoluminescence of the monolayer and hetero-bilayer regions in contact with the substrate greatly reduces background, improving nanobubble's PL contrast.³²

Figure 3a shows an AFM image of a nanobubble $\text{WSe}_2/\text{MoSe}_2$ on TS-Au that shows localized interlayer exciton

emission (**Figure 3b**) when compared with the flat heterostructure. The greater strength of the interlayer exciton emission on the nanobubble edge can be attributed to two factors. First, there is an enhancement from the strain of the nanobubble itself, which shifts the absorption of both constituent layers to the red, allowing for greater absorption of the excitation photons. Second, recent work has shown that, like their intralayer cousins, the interlayer excitons transition dipole is primarily in the plane of the 2D layers.³³ As the nano-optical probe moves across the edge of the nanobubble, more of the transition dipole is aligned with the polarization of the nano-optical field, allowing for significantly greater in- and out-coupling for optical fields. This alignment we believe is critical for near-field observation of interlayer exciton luminescence, which has substantially weaker oscillator strength. We also expect that strain-localized PL signal is enhanced by funneling effects, where excitons are preferentially shuttled toward the lower energy states.^{34–36}

Figure 3c shows point spectra at various locations across the nanobubble edges, identified by the colored markers in **Figure 3a,d**. Black arrows show visual identification of the localized interlayer exciton peak. Clear redshifting of the emission is observed as one moves from the outer edge of the bubble into the interior. To visualize this more clearly, in **Figure 3c** we plot a hyperspectral line scan along the vector defined by colored points in **Figure 3a**. The general trend is toward redder emission, shifting 200 meV over 15 nm into the bubble, after which the emission quickly falls to zero in the nanobubble

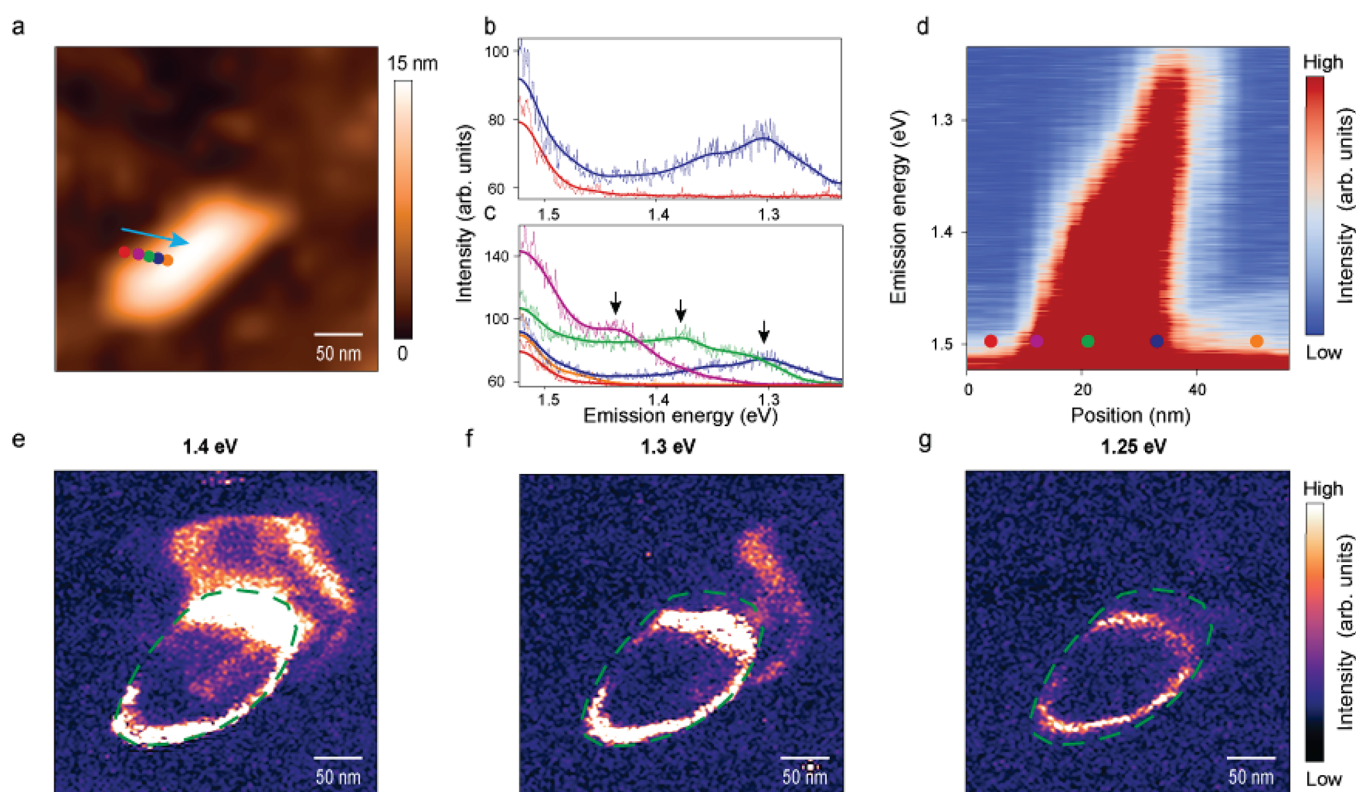


Figure 3. Hyperspectral nano-PL maps of localized interlayer exciton spectra. (a) Atomic force micrograph of MoSe₂–WSe₂ nanobubbles on a template-stripped Au substrate. (b) Average nano-PL spectra on (blue) and off (red) the nanobubble region. (c) Nano-PL point spectra across the bubble edge corresponding to the colored points in (a). For points farther into the interior of the nanobubble, the emissions energy redshifts. (d) Hyperspectral heat map along the vector defined by points in (a). The redshifting of the emission is clearly seen, with the spectral weight shifting from above 1.5 eV at the very edge of the bubble (red point) to below 1.3 eV at 35 nm inside the bubble (blue point). Past the position past 40 nm (orange point) the intensity shift below the low-energy detection cutoff of the EMCCD of 1.2 eV. (e–g) Nano-PL maps (average with a window size of 2.5 nm) binned different energy intervals, ordered by decreasing emission energy. The approximate physical nanobubble edge is shown with the green dashed line. The data clearly show the redshifting of the emission on going into the bubble.

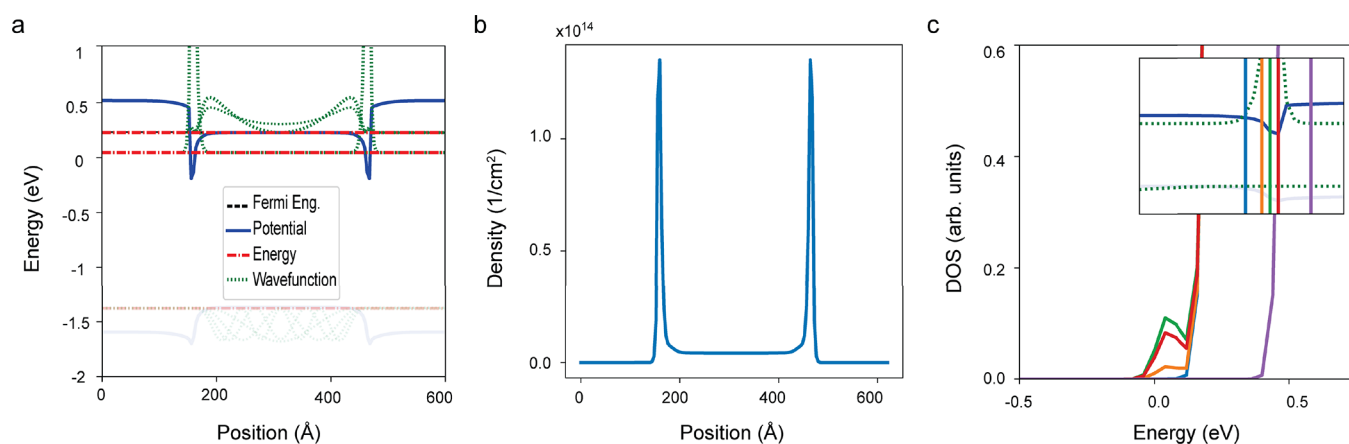


Figure 4. SP simulations. (a) Band diagram for 2D SP model of the bubble. Blue solid lines are the self-consistent band potentials, red dot-dashed lines are the energies of some of the confined states, black dashed line is the Fermi level, and green dotted lines are the confined wave functions. (b) Self-consistent electron density vs x position in (a). (c) DOS vs energy including the 1D states confined to the well, with 2D DOS outside of the bubble and in the bubble interior, superimposed.

interior. In Figure 3e–g we show this same trend with spatial maps with energy bins of 1.40, 1.30, and 1.25 eV, respectively. The size of the localized interlayer exciton emission “ring” clearly shrinks for the redder energy bins.

The shifting of the localized interlayer exciton emission energy reflects the energetic shift of the MoSe₂ conduction band observed in the scanning tunneling spectroscopy (STS).

Indeed, comparing the STS and nano-PL hyperspectral line scans, the roll off of the interlayer emission energy and the conduction band shows remarkable similarities at the nanobubble, but the nano-PL shows a sudden drop-off in emission intensity soon after the edge, which is not seen in STS. This drop-off however is expected due to the silicon detector used for the nano-PL spectroscopy (see Methods for details), which

rapidly loses quantum-efficiency at energies lower than ~ 1.3 eV. Similarly, we do not observe emission from the deeply localized defect states seen in Figure 2 given that this would correspond to emission energies of ~ 0.7 eV, which is far below the cutoff of our detector. While quantitative comparison between the STM and nano-PL data is not possible due to the sample differences, evaluation of the STS-derived band gap and exciton PL energy provides information on the binding energy for excitons associated with the conduction band near the bubble edge. Our results are consistent with a conduction band interlayer exciton binding energy with magnitude of a couple hundred milli-electronvolts within the bubble.³⁷

To understand the contributions of doping and strain to the observed spectroscopic features in experiment, we performed Schrödinger-Poisson (SP) simulations using the approach and code of ref 38. The length scale associated with the entire bubble region is too large to model directly, but we will show that the pertinent experimental features can be captured with a significantly simplified model. Our focus is on the conduction band of the heterostructure, which is derived from the MoSe₂ layer. Thus, we take band parameters for MoSe₂ from ref 39 and electromechanical parameters from ref 40. Since the important physics is localized to the bubble edge, we approximate the bubble region by a 2D inclusion in monolayer MoSe₂ (large enough to isolate the two interfaces required by periodic boundary conditions) with a downward shift of the conduction band in the bubble region as well as a background doping, which both increase with strain.⁴¹ The strain is assumed to be slightly larger at the edge of the bubble region. The density of states (DOS) is calculated assuming a 2D parabolic dispersion in the direction parallel to the inclusion above the conduction-band minimum and below the valence band maximum.

Figure 4a shows the band diagram obtained by self-consistent solution of the SP equations, focusing on the CB. By construction, the Fermi level is at the conduction band within the bubble region due to the background doping. The dips in the electrostatic potential of the conduction band at the edges of the “bubble” have two origins. The first is the band bending induced by the interface between the doped and undoped regions. The second is that we assume a higher strain at the interface, which increases the effect of this band bending. We found that adding a piezoelectric polarization charge at the interface (not shown) did not change the qualitative features, other than introducing an asymmetry in the band-bending on the opposite sides of the bubble region.

The red dot-dashed lines in Figure 4a are the energies of the bound states in the bubble region, and the green dotted lines are their squared wave functions (shifted so that the zero-density level is at the energy for clarity). We can see that the band bending at the interface region results in a bound state, which can also be clearly seen in Figure 4b as spikes in the electron density at the edge of the bubble regions. In Figure 4c we plot the DOS of the total system, including the sum of one-dimensional (1D) states in the bubble region and the 2D DOS in the bulk, at different x points near the interface of the bubble (see the inset, denoted by curve color). Superimposed on the step-like increase from the 2D DOS at the conduction and valence band edges in the bubble, we can see a small enhancement in the DOS near the interface, where the localized state resides. As we move toward the center of the bubble region, this enhancement weakens and, ultimately, disappears. We focused our model on the MoSe₂ layer of the

bilayer structure since the MoSe₂ is the main contributor to the conduction band. The size of the bubble is much larger than the crystal cell. If you were to zoom in at the atomic level the edge of the bubble can be approximated by its tangent on the surface of the MoSe₂ layer. Hence using the deformation gradients as a baseline for the energy of the different strained portions we can accurately model the band edge with the one-dimensional model. Our theoretical results, though the result of a simplified model, accurately capture the essential features of the experimental observations. They indicate that sharp lateral junctions in doping are an excellent way to engineer localized states in 2D TMD semiconductors. Such localized states do not depend on the presence of specific chemical defects or specialized strain fields. Recently, several 2D material interfaces have shown the ability to realize large charge transfer at the interface.^{42,43} Our results indicate that such interfacial engineering together with nanostructuring can be employed to create optical emitters with arbitrarily desired shapes at the nanoscale. Our work provides a clear route to achieving this in the future.

■ ASSOCIATED CONTENT

Data Availability Statement

The data used in this paper are available from the corresponding authors upon reasonable request.

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.2c02265>.

Experimental and theoretical methods; large-scale spectroscopic maps on the nanobubble; STM and nano-PL measurements on different bubbles; interlayer exciton emission energy (PDF)

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Author Contributions

S.S. carried out the STM/STS measurements and analysis. T.D. conducted optical measurements. T.D. and S.S. analyzed the nano-optical data. W.W. and E.Y. fabricated the device and performed SHG experiments. C.G. performed the theoretical calculations. J.H., X.Z., C.E.D., P.J.S., and A.N.P. provided advice. All authors contributed to writing the manuscript.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The single-crystal synthesis and heterostructure fabrication for this work was supported by the NSF MRSEC program through Columbia in the Center for Precision-Assembled Quantum Materials (PAQM), Grant No. DMR-2011738. STM measurements were supported by the NSF GOALI program under Grant No. DMR-1809122 and by the Air Force Office of Scientific Research via Grant No. FA9550-16-1-0601. C.E.D. and C.G. acknowledge support from the National Science Foundation under Grant No. DMR-1918455. The Flatiron Institute is a division of the Simons Foundation. P.J.S., T.D., and E.Y. acknowledge support from the National Science Foundation through Award No. DMR-2004437.

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