Semiconducting transition metal dichalcogenides (TMDCs), including MoS₂, MoSe₂, WS₂, and WSe₂, constitute a compelling platform for light–matter coupling at the single-quantum level. The unusually strong excitonic interactions in monolayers of these materials yield THz intrinsic radiative decay rates for spin-forbidden transitions and facilitate strong light–matter coupling, as has been demonstrated in MoSe₂. The van der Waals (vdW) bonding of layers of these materials further enables novel ways to create quantum emitters (QEs) through deposition of the materials on spatially aligned stressors, facilitating integration of QEs with photonic structures. Additionally, the spin-locked, valley-contrasting optical selection rules in TMDCs offer a route to realize spin-valley qubits for resident electrons.

Despite these desirable properties, all TMDC QEs reported to date are associated with nominally spin-forbidden transitions of dark excitons, as indicated by their large g factors under a perpendicular magnetic field. The corresponding optical transitions are relatively weak, with measured radiative rates of ~0.01 ns⁻¹ as discussed below, even with the likely brightening of the QEs associated with localization.

Here we address these issues by demonstrating engineered QEs in monolayer MoSe₂, a 2D TMDC semiconductor in which the lowest-lying exciton emits through a spin-allowed transition. Advancing previous works to create QEs in localized strain fields, we produce QEs in the material by patterning the substrate with nanoscale depressions, which can be created at desired spatial locations. According to our calculation, the resulting QEs promise radiative emission rates exceeding those of WSe₂ QEs by 2 orders of magnitude.

Our approach for creating exciton traps with controllable dimensions and placement relies on strain control. This method, as shown schematically in Figure 1a, complements the widely applied approach of creating localized strain by depositing the semiconductor monolayer on a protrusion in the substrate. We rely instead on the creation of milled depressions in the substrate but initially remain suspended above the indentation. This scheme has the advantage of avoiding tearing or wrinkling of the monolayer, which can be encountered during deposition of the monolayer on the substrate. We rely instead on the creation of milled depressions in the substrate, which can be encountered during deposition of the monolayer, which can be encountered during deposition of the monolayer.
respectively. Note the reduced line widths in part b vs part c.

sample, showing exciton and trion peaks at 1.66 and 1.63 eV, unstrained monolayer bandgap (at 2.33 eV) from a distance. (c) Reference PL spectrum (also at 5 K) excited above the deposition of the monolayer (Methods).

technique to prevent trapping of water at the interface during indentation for traps of

consequently produces an exciton confinement potential.

We construct the trap topography to mimic the shape of a clamped circular membrane under pressure (Supporting Information 1). Our method allows systematic control of exciton localization in potentials of varying size, unexplored in previous works. We can maintain the same depth of the confinement potential by scaling the width and depth of the trap proportionally. The smallest traps in this study are 50 nm in diameter and 5–6 nm deep. We utilize a novel transfer technique to prevent trapping of water at the interface during deposition of the monolayer (Methods).

We see clear signatures of exciton confinement for traps of various diameters (250, 120, and 50 nm). Low-temperature photoluminescence (PL) spectra for 250 nm traps (Figure 1b,c) illustrate the corresponding red shift of the exciton emission features. The neutral exciton and trion peaks found, respectively, at 1.66 and 1.63 eV for the unstrained monolayer are red-shifted by 80 meV, as expected for a strain of 1.5% (see Supporting Information 2 for the additional results for 250 and 120 nm structures). Strain confinement also reduces the emission line width. From no confinement to the 250 nm to 120 nm traps, the full width at half-maximum (fwhm) of the emission peak decreases from 8.5 to 4.0 to 1.5 meV (Figure 1b and Supporting Information 2). We ascribe this effect primarily to reduced inhomogeneous broadening, which has been shown to be the dominant line-broadening mechanism in previous studies employing four-wave mixing techniques.25 For the narrow lines discussed below, we ascribe the decreasing line width also to reduced lifetime broadening of the localized excitons.

For the case of 50 nm confinement, we see further significant reduction in the exciton line width, and we enter the regime of quantum emission. For this case, we have investigated an array of traps prepared on a grid, as shown in the atomic force microscope (AFM) topography and the spectrally filtered PL maps in Figure 2a,b. The PL spectrum from the confined excitons in trap QE1 is compared to that from free excitons in the unstrained region of the MoSe₂ monolayer in Figure 2c. The observation of a sharp, isolated emission line from the confined excitons differs from previous reports on a high background or clusters of peaks.26,27 We attribute the difference to our lithographically controlled strain, which produces a well-defined potential at a given position, yielding a well-defined and spectrally shifted emission feature.

Exciton emission spectra for the four 50 nm traps are presented in Figure 2d. From QE1 to QE3, line widths as low as 0.16 meV are observed. Similar sharp lines were also seen in another sample from several adjacent traps (not shown). Data acquired over long collection times exhibit broadening from spectral wandering of the emission line by ~0.5 meV (Supporting Information 3). We did not further study E4 because of its broad line width. The observed line widths of the confined excitons lie significantly below the radiative width of 1.4 meV for free excitons in MoSe₂. The reduced radiative rate for the confined excitons reflects the broad distribution of their center-of-mass (COM) momentum, which extends well beyond the light cone for photons in free space.29,30 We note that the measured PL lifetime of 0.19 ns for the confined excitons (Figure 2e) significantly exceeds the corresponding 2 ps lifetime of the free excitons at a similar temperature, although this comparison is complicated by the presence of nonradiative decay processes.

We confirm the single-photon emission character of the sharp peaks using a Hanbury Brown and Twiss (HBT) antibunching measurement. The coincidence dip was difficult to capture in a cw experiment because of the short exciton emission time but was clearly observable using picosecond pulsed excitation at a repetition rate of 75.8 MHz by binning the coincidence counts in 13.2 ns intervals corresponding to the timing of the excitation pulses. Figure 2f shows, for example, the integrated coincidence counts from QE1 in 13.2 ns bins. Comparing the zero-delay dip with the integrated counts at delays up to 66 ns yields g(2)(0) = 0.29 ± 0.13 (Methods). QE2 and QE3 had g(2)(0) values of 0.28 ± 0.12 and 0.31 ± 0.15, respectively (Supporting Information 4). The measured values below 0.5 unambiguously confirm the quantum emission character from these three traps. The nonzero value observed for g(2)(0) was limited by background light (Figure 2d) and by the pixels’ spectral wandering out of the detection bandwidth (Supporting Information 3).

To elucidate further the properties of these quantum emitters, we studied the influence of magnetic fields. Using
the Faraday geometry (magnetic field normal to monolayer) for the Zeeman measurements, we find that the single sharp emission peak bifurcates, as shown in Figure 3a for QE2. A plot of peak positions (Figure 3b) reveals a linear peak splitting, with a g factor of 4.08 ± 0.04, and with 95% confidence, a zero-field splitting ZFS < 60 μeV. A value of g ≈ 4 was observed for all of the measured QEs (Supporting Information 5), consistent with previous reports for free31,32 and localized36,27 excitons in MoSe2. This result confirms our assignment of the QEs as confined spin-allowed bright excitons. Note that the nominally spin-forbidden QEs in WSe2 exhibit a g-factor in the range of 6–102378,6,14 (Supporting Information 6).

We also measured the polarization characteristics of the PL to probe the anisotropy of confinement. Among all of the QEs, we observed less than 7% of linear PL polarization anisotropy at zero magnetic field (Figure 3c), suggesting azimuthal symmetry of the confining potential. This contrasts with earlier reports of linearly polarized PL from sharp peaks in MoSe2.27 We also show the circular polarization measurements in Supporting Information 5.

Regarding the origin of quantum emission in our 50 nm structures in MoSe2, strain confinement is clearly necessary. The unstrained regions of the MoSe2 monolayer produce only a weak, broad emission tail in the relevant spectral region (Figure 1b). However, the exciton interaction energy in the ideal 50 nm trap may be insufficient to dominate the experimental line widths (Supporting Information 7) as required to achieve quantum emission. Thus, we hypothesize that our QEs may benefit from potential fluctuations arising from variations in the local topography or other substrate characteristics, such as local trapped charges33 (see Supporting Information 8 for additional contributions to exciton localization and Supporting Information 10 for their implications on QE radiative rates). Biexciton states, which have been reported to be stabilized by a 20 meV binding energy in MoSe2,34 may also contribute to the presence of quantum emission. The importance of such localized biexcitons would depend on their rate of formation, which has not yet been established.

Our QEs could originate from the confinement of either bright neutral or charged exciton species.31,32 Both species are compatible with our experimental observations, including the measured value of g ≈ 4 (Supporting Information 9). Devices with electrical gating should not only elucidate this issue but also allow the controlled creation of either neutral or charged QEs. Indeed, both neutral and charged QEs provide new possibilities. Confinement of neutral excitons in MoSe2 is of interest as a platform to create single intervalley biexcitons for the generation of entangled photon pairs,35 since they suffer less from the complications found in WSe2 where the QEs exhibit spectral doublets3,4,6,12,13 and leak which-path information through photon energy and polarization. Confin-
azimuthal symmetry of QE spectrally integrated PL intensities for the various collection corresponding to the average of the V- and H-peak energies. The were performed at 1.8 K.

nominally spin-forbidden transitions have to date prevented ° for D (45 

for excitons in con have calculated (Supporting Information 10) the radiative rate inSupporting Information 10).

development of charged excitons, on the other hand, would permit the additional resident charge to be employed as an optically addressable spin-valley qubit. The slight difference in peak energy between the V- and H-spectrum is not due to zero-field splitting but rather to spectral wandering, as the spectrum for D (45°) linear polarization does not show a peak energy corresponding to the average of the V- and H-peak energies. The spectrally integrated PL intensities for the various collection polarizations show a degree of polarization less than 7%, suggesting azimuthal symmetry of QE’s confining potential. All measurements were performed at 1.8 K.

Figure 3. (a) The PL spectra of QE2 as a function of the strength of an applied magnetic field perpendicular to the sample. We attribute the high-energy tails in the spectra to spectral wandering as opposed to further splitting. (b) The extracted central energies from the QE2 doublet fitted linearly (red line) with respect to the magnetic field, revealing a g factor of 4.08 ± 0.04. (c) Polarized PL spectra from QE2 when the 1.62 eV laser excitation was vertically polarized. The slight difference in peak energy between the V- and H-spectrum is not due to zero-field splitting but rather to spectral wandering, as the spectrum for D (45°) linear polarization does not show a peak energy corresponding to the average of the V- and H-peak energies. The spectrally integrated PL intensities for the various collection polarizations show a degree of polarization less than 7%, suggesting azimuthal symmetry of QE’s confining potential. All measurements were performed at 1.8 K.

Looking forward, we estimate that the MoSe2 QEs radiate 100 times faster than QEs in WSe2 monolayers. Following established theories, describing the effect of localization, we have calculated (Supporting Information 10) the radiative rate for excitons in confinement potentials of varying size (Figure 4). Based on the intrinsic 2 ps⁻¹ radiative rate for the free exciton in monolayer MoSe₂, we find that localized excitons in MoSe₂ radiate faster than 1 ns⁻¹ for all plausible confinement scenarios (Supporting Information 10). Direct measurements on the QEs’ radiative rates through their quantum yields, however, were complicated by the aforementioned spectral wandering effect (Supporting Information 3): When excited with higher power toward saturation, the QEs generated more photons but also wandered and broadened, impeding isolation of single photons (see the Experimental Considerations section in Supporting Information 10).

By way of comparison, QEs in WSe₂ exhibit a radiative rate of just ~0.01 ns⁻¹. This value is inferred from the experimentally measured PL decay rates (0.07 ns⁻¹) adjusted for quantum yields (16.5%) (Supporting Information 6) and remains low even with probable brightening of the QEs attributed to defect-induced broken valley symmetry. The low radiative rates of existing QEs in TMDC systems with nominally spin-forbidden transitions have to date prevented access to the regime of strong coupling to optical cavities. As the spin-allowed QE transitions in MoSe₂ possess estimated radiative rates comparable to those of self-assembled InAs/GaAs quantum dots, they hold the potential not only for high-speed communication systems but also for single-photon nonlinearity facilitated by entering the strong coupling regime.

■ METHODS

Sample Fabrication and Characterization. We have applied a lithographically defined method of strain control to realize exciton confinement in monolayer MoSe₂. Our MoSe₂ monolayers were exfoliated from commercial crystals (HQ Graphene) grown by chemical vapor transport. The monolayers were deposited on a silicon substrate with an oxide (SiO₂) overlayer in which traps with the desired curved topography had been milled in the substrate using a focused ion beam (FIB, FEI Helios NanoLab 600i). The subsequent pressurization of monolayers was achieved by a polymer molding technique that we developed: After a monolayer was deposited on the patterned substrate using polydimethylsiloxane (PDMS) or polycarbonate (PC) as the polymer for transfer, the polymer film, initially suspended above the indentation, was molded by thermal stress to pressurize the attached monolayer. To prevent trapping of water at the interface during the transfer process, we kept the substrate at temperatures above 120 °C prior to and during the monolayer deposition on the substrate. The polymer film was eventually removed by dissolving it in chloroform. While only cryogenic measurements are shown in this manuscript, we verified that our strain method with polymer molding, or with other pressurization techniques including gas pressurization and nanoindentation, can routinely produce biaxial strain greater than 1% in monolayers (unpublished experiments). The nanoscale topography of the samples was measured using an atomic-force microscope (AFM, Park NX-10) in the non-contact mode with hardened probes (MikroMasch HQ:NSC15/Hard/Al BS).

Low-Temperature Photoluminescence Microscopy. Low-temperature PL was performed in a top-loading cryostat with a superconducting magnet (Attocube attoDRY2100).
Excitation and collection light was defined with an x–y scanning mirror (Newport FSM-300) mounted above the cryostat and focused onto the sample using a cryogenic objective (Attocube LT-APO/VISIR/0.82) inside the cryostat. Below-gap excitation was provided with a tunable pulsed Ti:sapphire laser (Coherent Mira 900) with a pulse width of 3 ps and a repetition rate of 75.8 MHz. The collected PL was separated from scattered light from the excitation beam using long-pass filters (Semrock 785RE) and subsequently dispersed in a spectrometer with 0.12 meV resolution (Princeton Instruments SP2300) using a 1800 g mm⁻¹ grating and a charge-coupled device (CCD, Andor Newton 940) for detection.

**Time-Correlated Single-Photon Counting.** Time-resolved PL and g²(2) correlation were obtained using a time-correlated single-photon counting (TCSPC, PicoHarp300) module. For photon counting, light from the QE was spectrally filtered down to a 0.5 meV bandwidth using a monochromator with a 1800 g mm⁻¹ diffraction grating (Newport 33067FL01-290R). For time-resolved PL, single photons were detected using a fast photon counter (~50 ps jitter, MPD PDM). The detector output, together with the trigger from the pulsed excitation laser, was recorded in the TCSPC module to obtain a histogram of photon arrival number as a function of time delay. From the histogram, the decay time was extracted. The instrument response function was obtained using the laser pulses, which yielded a fwhm <64 ps. For g²(2) correlation measurements, a Hanbury Brown and Twiss (HBT) setup was employed. Single photons were split in a fiber beamsplitter, detected using two single photon counting modules optimized for detection efficiency (PerkinElmer SPCM-AQR14), and recorded using the TCSPC module in the time-tagged time-resolved (TTTR) mode. To mitigate the spectral wandering of the QE's (Supporting Information 3), we kept the excitation power very low (<30 nW). The resulting count rate on the SPCMs was around 2000 s⁻¹, and the total effective integration time for each QE was 4 h on average. We also measured the QE's full spectra to center our monochromator prior to starting every 60 min TTTR integration and used the spectra to extract the QE's where the QEs wandered significantly (Supporting Information 3). The recorded photon streams were processed to construct a histogram of photon coincidences as a function of time delay. From the histogram, we obtained the integrated coincidence counts (as shown in Figure 2f) by summing up the coincidences in 1.2 ns wide time windows centered around the periodic laser excitation. The 1.2 ns width was approximately the fwhm of the instrument response function when using SPCMs with a timing jitter greater than 600 ps. The g²(0) values were calculated from the integrated coincidence counts in the time-zero delay peak divided by the average of the adjacent eight effective peaks (Supporting Information 4). The standard deviation of the g²(2)(0) measurement was obtained from the propagated Poissonian counting statistics of the integrated coincidence counts.

**ASSOCIATED CONTENT**

1. **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c04282.

Strain distribution in traps, additional experimental results on exciton emission for traps of various diameters, spectral wandering of exciton emission, analysis of g²(2) correlation data, additional QE magneto-optical and polarization measurements, characteristics of WSe₂ quantum emitters, modeling of exciton-localization effects in traps, additional contributions toward exciton localization in MoSe₂ quantum emitters, distinguishing between neutral excitons and charged excitons, radiative rates of localized excitons, and temperature-dependent photoluminescence from MoSe₂ quantum emitters (PDF)

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**Author Contributions**

M.D. and J.L.Z. contributed equally. L.Y. and T.F.H. conceived the project. L.Y. and M.D. fabricated the samples. L.Y., M.D., and J.L.Z. performed the optical measurements. L.Y. and J.L.Z. compiled the instrumentation control. L.Y. and S.B. analyzed the data including the TTTR photon stream. B.K., J.V., and T.F.H. supervised the project. L.Y. wrote the paper with input from all authors.

**Notes**

The authors declare no competing financial interest.

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