Large-scale quantum-emitter arrays in atomically thin semiconductors

Carmen Palacios-Berraquero1,*, Dhiren M. Kara1,*, Alejandro R.-P. Montblanch1, Matteo Barbone1,2, Pawel Latawiec3, Duhee Yoon2, Anna K. Ott2, Marko Loncar3, Andrea C. Ferrari2 & Mete Atatüre1

Quantum light emitters have been observed in atomically thin layers of transition metal dichalcogenides. However, they are found at random locations within the host material and usually in low densities, hindering experiments aiming to investigate this new class of emitters. Here, we create deterministic arrays of hundreds of quantum emitters in tungsten diselenide and tungsten disulphide monolayers, emitting across a range of wavelengths in the visible spectrum (610–680 nm and 740–820 nm), with a greater spectral stability than their randomly occurring counterparts. This is achieved by depositing monolayers onto silica substrates nanopatterned with arrays of 150-nm-diameter pillars ranging from 60 to 190 nm in height. The nanopillars create localized deformations in the material resulting in the quantum confinement of excitons. Our method may enable the placement of emitters in photonic structures such as optical waveguides in a scalable way, where precise and accurate positioning is paramount.
transition metal dichalcogenides (TMDs) are optically active, semiconducting layered materials (LMs) which can be exfoliated down to monolayers. These are of particular interest, as they exhibit properties such as an optically accessible valley degree of freedom that is locked to the exciton spin\(^1\)\(^{\text{-}}\)^\(^3\), strong two-dimensional confinement, favouring bound excitonic states\(^4\)\(^{\text{-}}\)^\(^5\) and the opportunity to investigate many-body physics\(^6\). Intrinsic properties of LMs—atomically precise interfaces, lack of dangling bonds, flexibility and the possibility of stacking different LMs into functional heterostructures\(^7\)\(^{\text{-}}\)^\(^8\)—make them not only interesting for fundamental physics but also suitable for technological applications\(^9\)\(^{\text{-}}\)^\(^{\text{10}}\). For these reasons, the identification of quantum emitters (QEs) in LMs\(^11\)\(^{\text{-}}\)^\(^{16}\) has generated much excitement in the field of two-dimensional nanophotonics\(^10\)\(^{\text{17}}\)\(^{\text{-}}\)^\(^{\text{19}}\) and quantum technologies\(^9\)\(^{\text{20}}\).

Single photon emission has been seen from QEs in tungsten diselenide (WSe\(_2\)) with both above bandgap\(^11\)\(^{\text{-}}\)^\(^{16}\) and resonant optical excitation\(^21\). In addition, QEs in WSe\(_2\) and tungsten disulphide (WS\(_2\)) have been implemented in heterostructures to achieve electrically driven single-photon emission\(^22\). However, the origin of QEs in TMDs is still unclear, and has been assigned to both defects\(^11\)\(^{\text{-}}\)^\(^{16}\) and strain gradients\(^21\)\(^{\text{-}}\)^\(^{24}\). Experiments on these QEs have, until now, been reliant on their rare and random occurrence. Deterministic creation of precisely positioned LM QEs in large numbers is important for accelerating the study of these emitters, as well as opening up the prospect for scalability and on-chip applications.

Here, we report a method to create arrays of single-photon emitting QEs in WSe\(_2\) and quantum-like emitters in WS\(_2\) using a nanopatterned silica substrate. We obtain structures with QE numbers typically in the range of hundreds. The quality of these deterministic QEs surpasses that of their randomly appearing counterparts, with spectral wanderings of \(\sim 0.1\) meV—an order of magnitude lower than previous reports\(^11\)\(^{\text{-}}\)^\(^{15}\). Our technique is a crucial first step towards solving the scalability challenge for LM-based quantum photonic devices.

**Results**

**Nanopatterned substrate preparation and characterization.** To create large-scale QE arrays in LMs, we place the active material on patterned structures fabricated on the substrate in order to create spatially localized physical disturbances to the otherwise flat LM flakes. To this end, we first pattern arrays of nanopillars of different heights, ranging from 60 to 190 nm, on silica substrates using electron beam lithography. Figure 1a shows a scanning electron microscope image of one such substrate of 130 nm nanopillar height. We place layers of WSe\(_2\) and WS\(_2\) on the nanopillars as follows. Bulk WSe\(_2\) and WS\(_2\) crystals are characterized before exfoliation as described in ref. 22. These are then exfoliated on a polydimethylsiloxane layer by micromechanical cleavage\(^7\)\(^{\text{-}}\)^\(^{25}\). Single-layer (1L) samples are identified first by optical contrast\(^26\), and the selected 1L-WSe\(_2\) and 1L-WS\(_2\) flakes are then placed onto the patterned nanopillar substrate via an all-dry viscoelastic transfer technique due to their higher adhesion to SiO\(_2\) (refs 7,19,27), as schematically shown in Fig. 1b. After exfoliation and transfer, the 1L-WSe\(_2\) and 1L-WS\(_2\) flakes are characterized by Raman spectroscopy\(^26\)\(^{\text{27}}\), photoluminescence (PL)\(^30\) and atomic force microscopy (AFM), confirming the transfer and that the process does not damage the samples (see Supplementary Fig. 1 and Supplementary Note 1 for the corresponding spectra and discussion). Figure 1c is an AFM scan of a 1L-WSe\(_2\) flake over a single nanopillar. The bottom panel of Fig. 1c plots the height profile of the 1L-WSe\(_2\) flake taken along the dashed pink line. This reveals how the flake (solid pink line) tent over the nanopillar. The blue-shifted area corresponds to the measured profile of a bare nanopillar. Figure 1d is a dark-field optical microscopy image of part of a 43,000 \(\mu\)m\(^2\) 1L-WSe\(_2\) flake on a substrate patterned with a 4-\(\mu\)m-spaced nanopillar array with nominal height of 130 nm. The regularly spaced bright spots correspond to nanopillar sites. We see locations providing brighter scattering (two examples are encircled in pink) and others showing fainter intensity (two examples are encircled in blue). By correlating with AFM measurements we find that the former correspond to locations where the 1L-WSe\(_2\) tents over the nanopillars and the latter correspond to locations where the flake is pierced by the nanopillars (see Supplementary Fig. 2). On average, we find that two-third of the sites are not pierced during the deposition step.

**Quantum light from WSe\(_2\)-based deterministic QEs.** Figure 2a is an integrated raster scan map of PL emission at \(\sim 10\) K of six adjacent non-pierced nanopillar sites in the region enclosed by the green dashed line in Fig. 1d. The most prominent feature is the \(\sim 10\) increase in intensity at the location of every nanopillar. Figure 2b reveals the source of this emission enhancement: spectra taken at each nanopillar location display bright sub-nanometre linewidth emission peaks. Figure 2c demonstrates the single-photon nature of this emission via photon-correlation measurements taken (from left to right) at the first, third and fourth nanopillar locations. Ten nanometre band-pass filters, indicated by the pink, green and blue highlighted areas in the panels of Fig. 2b, select the spectral windows for the photon-correlation measurements. We obtain \(g^{(2)}(0)\) values of 0.0868 ± 0.0065, 0.170 ± 0.021 and 0.182 ± 0.028, respectively, uncorrected for background emission or detector response. While these surpass those in early reports\(^11\)\(^{\text{-}}\)^\(^{15}\), we expect the quality of the single-photon emission from the QEs to improve under resonant excitation\(^21\). Out of the 53 unpierced nanopillar sites in this substrate we find sub-nm emission peaks in 51 of them, giving \(\sim 96\%\) yield in QE generation. Their emission wavelength ranges between 730 and 820 nm (see Supplementary Fig. 3 for statistics), equivalent to a redshift distributed between 50 and 280 meV from the unbound exciton emission energy at \(\sim 1.755\) eV (ref. 32), as observed for the naturally occurring QEs in WSe\(_2\) (refs 11–15). The fine-structure splitting (200–730 \(\mu\)eV) and the emission linewidths as narrow as \(\sim 180\) \(\mu\)eV (\(\sim 0.08\) nm) are also consistent with previous reports\(^11\)\(^{\text{-}}\)^\(^{15}\) (Supplementary Fig. 3) advocating that these deterministically created QEs are of the same nature as the randomly appearing ones.

**Effect of nanopillar height on WSe\(_2\)-based QEs.** To study the effect of nanopillar height, we carry out similar optical measurements of 1L-WSe\(_2\) flakes deposited on nanopillars of height \(\sim 60\) and \(\sim 190\) nm. The spectra taken at the 60-nm nanopillars have multiple peaks of \(\sim 1\) nm linewidth on average (see Supplementary Fig. 3 for example spectra). In contrast, Fig. 2d is a representative spectrum taken from the 190 nm nanopillars, displaying a better isolated, single sub-nm emission peak. The inset reveals a 722 meV (ref. 32), as observed for the naturally occurring QEs in WSe\(_2\) (refs 11–15). The fine-structure splitting (200–730 \(\mu\)eV) and the emission linewidths as narrow as \(\sim 180\) \(\mu\)eV (\(\sim 0.08\) nm) are also consistent with previous reports\(^11\)\(^{\text{-}}\)^\(^{15}\) (Supplementary Fig. 3) advocating that these deterministically created QEs are of the same nature as the randomly appearing ones.
increased. For the 190 nm nanopillars, 50% of all nanopillar sites host a single QE with one emission peak, as indicated by the purple bars. Spectral wandering of the peaks as a function of time also displays a strong dependence on the nanopillar height. To quantify this dependence, we record the maximum range of emission wavelength wandering per QE over tens of seconds. The solid black circles in Fig. 2f correspond to the mean of these values for each group of QEs pertaining to each nanopillar height, for 17 different QEs in total, with the error bars displaying the standard deviation of these distributions. We observe a reduction from a few meV for 60 nm nanopillars to below 0.25 meV (average) for the tallest 190 nm nanopillars (see Supplementary Fig. 4), reaching as low as 0.1 meV. To the best of our knowledge, this is the lowest spectral wandering seen to date in LM QEs11–15. Hence, these deterministic QEs are comparable, and even superior, in spectral stability to their randomly appearing counterparts. The dependence of certain QE characteristics on nanopillar height, along with shifts in the delocalized neutral exciton peak (X0) at room temperature30 at the nanopillar locations (see Supplementary Fig. 5), suggest that a localized strain gradient induced by the nanopillars might be playing an active role in producing QEs, as well as determining their specific optical properties21,23,24.

**Deterministic QE creation in WS2 monolayers.** The method we present for QE creation is not restricted to a specific LM. We predict a similar effect on different LMs and test this by using 1L-WS2. Despite previous efforts to measure QEs in 1L-WS2, there has only been one previous report of single-photon emission in this material22. Figure 3a shows an integrated PL intensity raster scan map taken at 10 K of a 1L-WS2 on a substrate with 170-nm-high nanopillars square array spaced by 3 μm. The inset shows a true-colour dark-field optical microscopy image of the same flake, where the red areas (due to fluorescence) are 1L-WS2. Once again, the brighter spots correspond to the unpierced nanopillar locations, as verified by AFM measurements, and show overlap with the bright fluorescence spots in the PL intensity image where, similar to WSe2, intensity is increased (here by a factor ~4) at every one of the 22 non-pierced nanopillar sites in the flake. Panel 1 of Fig. 3b shows the typical 1L-WS2 emission spectrum at ~10 K (ref. 22), measured from a flat region of the same flake away from the nanopillars. The X0 and X− unbound
Excitons are labelled in the figure, while the broad red-shifted emission band arises from weakly localized or defect-related excitons in the 1L-WS$_2$ at low temperatures, and is present in this material regardless of location. Panels 2 and 3 of Fig. 3b show representative PL emission spectra taken at nanopillars of heights $B_{170}$ and $B_{190}$ nm, respectively, where once again sub-nm spectral features arise. We note that we observe fine-structure splitting for WS$_2$ in these QEs, which range from 300 to 810 meV (Supplementary Fig. 6), as represented in the panel insets corresponding to the spectral regions highlighted in red. We also measure the spectrum of several WS$_2$ QEs as a function of time (see Supplementary Fig. 6) and find all spectral wandering values below 0.5 meV over 1–2 min. Figure 3c shows statistics on QE emission wavelength collected for over 80 QEs for 1L-WS$_2$ on 170-nm (white bars) and 190-nm (red bars) nanopillars. The wavelength distribution of the sub-nm emission lines, typically in the 610–680 nm region (53–300 meV redshift from X$_0$), is as narrow as 20 nm for the 190-nm nanopillars. Most nanopillar sites on WS$_2$ show multiple sub-nm lines, suggesting the creation of several QEs at each site for these nanopillar heights. Figure 3d plots a histogram of the number of sub-nm peaks appearing at each nanopillar for both nanopillar heights. The trend is similar to that seen in WSe$_2$, where higher nanopillars lead to a narrower spread in the number of peaks towards a higher likelihood of

Figure 2 | Creation of quantum emitter arrays in 1L-WSe$_2$. (a) Integrated PL intensity raster scan of the region enclosed by the green rectangle in Fig. 1d, taken under 200 nW $\mu$m$^{-2}$, 532 nm laser excitation at 10 K. Green crosses mark the position of the six nanopillars beneath the 1L-WSe$_2$. Colour-scale bar maximum, 160 kcounts s$^{-1}$. (b) PL spectra taken at each of the corresponding green crosses in a, from left to right respectively, showing the presence of narrow lines at each nanopillar location. (c) Photon correlation measurements corresponding to the filtered spectral regions (10 nm wide) enclosed by the blue, green and pink rectangles, in b, with $g^{(2)}(0) = 0.087 \pm 0.065$, 0.17 $\pm$ 0.02 and 0.18 $\pm$ 0.03, and rise times of 8.81 $\pm$ 0.80 ns, 6.15 $\pm$ 0.36 ns and 3.08 $\pm$ 0.41 ns, respectively. (d) Spectrum taken from a 1L-WSe$_2$ on a 190 nm nanopillar, showing lower background and a single sub-nm emission peak. Higher-resolution spectrum in the inset reveals the fine-structure splitting of this QE peak. An asymmetry can be seen in the spectrum, which has been previously attributed to a phonon sideband in naturally occurring QEs. (e) Probability distribution (in %) of the number of emission lines per nanopillar for samples using different nanopillar heights (60, 130 and 190 nm in white, blue and purple, respectively). A trend of higher probability of single QE emission peaks per nanopillar location with increasing height is evident, reaching 50% for 190 nm nanopillars. (f) Increasing nanopillar height also leads to a reduction of spectral wandering. Solid black circles represent the mean value of spectral wandering of several QEs for a given nanopillar height, while the error bars represent the standard deviation of each distribution, both extracted from time-resolved high-resolution spectral measurements (Supplementary Fig. 4). A total number of seven samples was used to collect the statistics necessary for Fig. 2e,f.
creating a single QE at each nanopillar site. We note that we obtain a 95% yield of QE creation in 1L-WS2 on non-pierced nanopillars. Further, ∼75% of these display two or less sub-nm emission peaks. In contrast, the 60- and the 130-nm-high nanopillars do not result in any QE occurrence (see Supplementary Fig. 7 for examples of these measurements).

Discussion
We presented a simple method for the deterministic creation of scalable arrays of quantum-light emitters embedded in LMs emitting at different regions of the optical spectrum33. The reliability of the technique will accelerate experimental studies of QEs in TMDs, which at present rely on their rather rare and random occurrence11–15. In the immediate future, a detailed study is necessary in order to achieve a better understanding of the specific role of nanopillar height and geometry in defining the characteristics of the quantum emission. We expect tunability of the optical emission by varying the shapes of the underlying nanostructures. This is important, for example, when considering inhomogeneous line-broadening due to charge noise. Further, the flexibility in the choice of substrate, in turn, provides an opportunity to create hybrid quantum devices where LM QEs can be coupled to quantum systems in other materials such as spins in diamond and silicon carbide.

Methods
Substrate preparation. The silica nanopillar substrate is fabricated with a high-resolution direct-write lithographic process via spin-on-glass polymer hydrogen silsesquioxane (HSQ)30. First, a wafer with 2 μm thermal oxide is cleaved and then cleaned. HSQ resist (FOXs-16, Dow-Corning) is diluted with methyl isobutyl ketone (MIBK) in different ratios and spun onto the substrate, giving variable thickness depending on the dilution. After baking at 90 °C for 5 min, the substrate is exposed in an electron beam lithography tool (Elionix F-125) and then developed in a 25% solution of tetramethyl ammonium hydroxide (TMAH) developer and rinsed in methanol. To convert the defined structures into pure SiO2, we apply rapid thermal annealing at 1,000 °C in an oxygen atmosphere37, resulting in arrays of sharply-defined sub-100 nm silica nanopillars.

Optical measurements. Room temperature Raman and PL measurements are carried out using a Horiba LabRam HR Evolution microspectrometer equipped with a ×100 objective (numerical aperture 0.9) and a spot size < 1 μm. The pixel-
to-pixel spectral resolution for the Raman measurements is ~0.5 cm$^{-1}$. Bragg gratings (BraggGrate) are used to detect the ultralow frequency Raman peaks. The power is kept below 50 µW to prevent heating effects. The excitation wavelength used is 314.5 nm for WSe$_2$ and 457 nm for WS$_2$.

A variable-temperature helium flow cryostat (Oxford Instruments Microstat HiRes2) is used to perform low-temperature PL measurements with a home-built confocal microscope mounted on a three-axis stage (Physik Instrumente M-400/DG) with a 5-cm travel range, 290-nm resolution for coarse alignment and a piezo scanning mirror (Physik Instrumente S-334) for high-resolution raster scans. PL is collected using a 1.7-mm working distance objective with a numerical aperture of 0.7 (Nikon S Plan Fluor ×60) and detected on a fibre-coupled single-photon-counting module (PerkinElmer: SPCM-AQRH). Photoemissions from a Hanbury Brown and Twiss interferometer are recorded with a time-to-digital converter (qTSEQ). A double grating spectrometer (Princeton Instruments) is used for acquiring spectra. For PL measurements, the excitation laser (532 nm, Laser Quantum) is suppressed with a long pass filter (550 nm Thorlabs FEL0550).

**Data availability.** The data that supports the findings of this study are available from the corresponding author upon request.

**References**

1. Cao, T. et al. Valley-selective circular dichroism of monolayer molybdenum disulphide. Nat. Commun. 3, 887 (2012).
2. Zeng, H., Dai, J., Yao, W., Xiao, D. & Cui, X. Valley polarization in MoS$_2$ nanolayers by optical pumping. Nat. Nanotechnol. 7, 490–493 (2012).
3. Mak, K. F., He, K., Shan, J. & Heinz, T. F. Control of valley polarization in atomically thin transition metal dichalcogenides. Nat. Mater. 12, 207–211 (2013).
4. He, K. et al. Tightly bound excitons in monolayer WSe$_2$. Phys. Rev. Lett. 113, 26803 (2014).
5. Sidler, M. et al. Fermi-polaron-polaritons in charge-tunable atomically thin semiconductors. Nat. Phys. 13, 255–261 (2016).
6. Bonaccorso, F. et al. Production and processing of graphene and 2d crystals. Mater. Today 15, 564–589 (2012).
7. Akinwande, D., Petrone, N. & Hone, J. Two-dimensional flexible nanoelectronics. Nat. Commun. 5, 5678 (2014).
8. Majumdar, A. et al. Hybrid 2D material nanophotonics: a scalable platform for low-power nonlinear and quantum optics. ACS Photonics 2, 1160–1166 (2015).
9. Xie, F., Wang, H., Xiao, D., Dubey, M. & Ramasubramaniam, A. Two-dimensional material nanophotonics. Nat. Photonics 8, 899–907 (2014).
10. Onndorf, P. et al. Single-photon emission from localized excitons in an atomically thin semiconductor. Optica 2, 347 (2015).
11. Srivastava, A. et al. Optically active quantum dots in monolayer WSe$_2$. Nat. Nanotechnol. 10, 491–496 (2015).
12. He, Y.-M. et al. Single quantum emitters in monolayer semiconductors. Nat. Nanotechnol. 10, 497–502 (2015).
13. Koperski, M. et al. Single photon emitters in exfoliated WSe$_2$ structures. Nat. Nanotechnol. 10, 503–506 (2015).
14. Chakraborty, C., Kunnischtk, L., Goodfellow, K. M., Beams, R. & Varnikav, A. N. Voltage-controlled quantum light from an atomically thin semiconductor. Nat. Nanotechnol. 10, 507–511 (2015).
15. Tran, T. T., Bray, K., Ford, M. J., Toth, M. & Aharonovich, I. Quantum emission from hexagonal boron nitride monolayers. Nat. Nanotechnol. 11, 37–41 (2015).
16. Bonaccorso, F., Sun, Z., Hasan, T. & Ferrari, A. C. Graphene photonics and optoelectronics. Nat. Photonics 4, 611–622 (2010).
17. Koppens, F. H. L. et al. Photodetectors based on graphene, other two-dimensional materials and hybrid systems. Nat. Nanotechnol. 9, 780–793 (2014).
18. Ferrari, A. C. Science and technology roadmap for graphene, related two-dimensional crystals, and hybrid systems. Nanoscale 7, 4898–4810 (2015).
19. Aharonovich, I., Englund, D. & Toth, M. Solid-state single-photon emitters. Nat. Photonics 11, 631–641 (2016).
20. Branny, A. et al. Discrete quantum dot like emitters in monolayer MoSe$_2$: spatial mapping, magneto-optics, and charge tuning. Appl. Phys. Lett. 108, 142101 (2016).
21. Palacios-Berraquero, C. et al. Atomically thin quantum light-emitting diodes. Nat. Commun. 7, 12978 (2016).
22. Koppens, F. H. L. et al. Photodetectors based on graphene, other two-dimensional materials and hybrid systems. Nat. Nanotechnol. 9, 780–793 (2014).
23. Ferrari, A. C. Science and technology roadmap for graphene, related two-dimensional crystals, and hybrid systems. Nanoscale 7, 4898–4810 (2015).
24. Aharonovich, I., Englund, D. & Toth, M. Solid-state single-photon emitters. Nat. Photonics 11, 631–641 (2016).
25. Branny, A. et al. Discrete quantum dot like emitters in monolayer MoSe$_2$: spatial mapping, magneto-optics, and charge tuning. Appl. Phys. Lett. 108, 142101 (2016).
26. Palacios-Berraquero, C. et al. Atomically thin quantum light-emitting diodes. Nat. Commun. 7, 12978 (2016).
27. Kumar, S. et al. Resonant laser spectroscopy of localized excitons in monolayer WSe$_2$. Optica 3, 882 (2016).
28. Wang, C. et al. Valley dynamics probed through charged and neutral exciton emission in monolayer WSe$_2$. Phys. Rev. B 90, 075413 (2014).
29. Jiang, M., Kurvits, J. A., Lu, Y., Nurmiokko, A. V. & Zia, R. Reusable inorganic templates for electrostatic self-assembly of individual quantum dots, nanodiamonds, and lanthanide-doped nanoparticles. Nat. Nanotechnol. 15, 5010–5016 (2015).
30. Kang, K. et al. High-mobility three-atom-thick semiconducting films with wafer-scale homogeneity. Nature 520, 656–660 (2015).
31. Ling, X. et al. Parallel stitching of 2D Materials. Adv. Mater. 28, 2322–2329 (2016).
32. Grigorescu, A. E. et al. Resists for sub-20-nm electron beam lithography with a focus on HSQ: state of the art. Nano Lett. 15, 9677–9683 (2015).
33. Holzwarth, C. W., Barwicz, T. & Smith, H. I. Optimization of hydrogen silsesquioxane for photonic applications. J. Vac. Sci. Technol. B Microelectron. Nanometer Struct. Process. Meas. Phenom. 25, 2658 (2007).

**Acknowledgements**

We acknowledge financial support from the Marie Sklodowska-Curie Actions Spin-NANO, Grant No. 676108, EU Graphene Flagship, ERC Grants Hetero2D and PHOENICS, EPSRC Grants EP/K01711X/1, EP/K017144/1, EP/N010345/1, EP/M057799/1, EP/I0116067/1, Quantum Technology Hub NQIT EP/M013243/1, the EPSRC Cambridge NanodTC, Graphene Technology CDT, EP/G037221/1 and the STC Center for Integrated Quantum Materials (NSF Grant No. DMR-1231319). We thank H.S. Knowles and P. Borisova for technical assistance.

**Author contributions**

M.A., A.C.F. and M.L. managed the project. M.A., D.M.K. and C.P.-B. devised the project. P.L. fabricated the nanopatterned substrates. C.P.-B., A.R.-P.M., D.M.K., M.A., A.C.F. and M.L. managed the project. M.A., D.M.K. and C.P.-B. developed the deposition method and fabricated the samples. P.L. fabricated the nanopatterned substrates. C.P.-B., A.R.-P.M. and M.B. developed the deposition method and fabricated the samples. C.P.-B., A.R.-P.M., D.M.K., A.K.O., M.B. and D.Y. performed the optical measurements. C.P.-B., A.R.-P.M. and M.B. performed AFM measurements. All authors participated in the results discussions, analysis and the writing of the manuscript.

**Additional information**

**Supplementary Information** accompanies this paper at http://www.nature.com/naturecommunications

**Competing interests:** The authors declare no competing financial interests.

**Reprints and permission information** is available online at http://npg.nature.com/reprintsandpermissions

**How to cite this article:** Palacios-Berraquero, C. et al. Large-scale quantum-emitter arrays in atomically thin semiconductors. Nat. Commun. 8, 15093 (2017). doi: 10.1038/ncomms15093

**Publisher’s note:** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.