Supporting Information for

Dual-Wavelength Lasing in
Quantum-Dot Plasmonic Lattice Lasers

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S1. Sample Fabrication

Materials. Four-inch diameter, single-side-polished, single-crystalline Si(100) wafers with <0.4 nm root-mean-square (RMS) roughness and thicknesses of 500 μm were purchased from Silicon Quest. Octadecyltrichlorosilane (ODTS, 96%) was purchased from Merck KGaA. Bicyclohexyl (99%, D79403), carbon tetrachloride (CCl4, 99.5%, 289116), chloroform (99.8%, 132950), and sulfuric acid (H2SO4, 95–98%, 258105) were purchased from Sigma. Hydrogen peroxide (H2O2, 30%, AnalR Normapur) was purchased from VWR. Nitric Acid (65% HNO3) was purchased from Fisher Chemical. 18.2 MΩ deionized water (DI) was obtained from a MilliQ Advantage A10 water-purification system. Silver (Ag, 99.999%) pellets were purchased from Kurt Lesker. Tungsten dimple boats (49 × 12 × 0.4 mm3) were purchased from Umicore. PF Gel-Films (X4, 6.5 mil) were purchased from Gel-Pak. Acetone (Technic France, Micropur VLSI Grade), isopropanol (IPA, Technic France, Micropur VLSI Grade), CSAR 62 resist (AR-P 6200.09, Allresist GmbH), and developer for CSAR 62 resist (AR 600-546, Allresist GmbH) were provided by the Binnig and Rohrer Nanotechnology Center (BRNC) at IBM Zurich.
**Fabrication of template for plasmonic lattice.** A Si(100) wafer with four-inch diameter was first coated with a 50-nm-thick SiO$_2$ layer using atomic layer deposition (Oxford Instruments, FlexAL, O$_2$ plasma at 200 °C, 0.8 sec pulse duration). The wafer was then diced into 1.5×1.5 cm$^2$ chips. Before further processing, the chips were cleaned using acetone, isopropanol, and O$_2$ plasma (PVA-TePla, GIGAbatch 310M, 600 W, 5 min). After a prebake (hotplate, 180 °C, 10 min), the chips were coated with a 127-nm-thick layer of AR-P 6200.04 (spin coater, 2000 revolutions per minute) and then soft-baked (hotplate, 150 °C, 5 min). Arrays of circles were defined into the resist by first using electron-beam lithography (Vistec, NFL 5) and then developing the sample in AR 600-546 for 1 min followed by rinsing in isopropanol and drying with a N$_2$ gun. The patterned resist served as a mask to selectively etch circular trenches into the SiO$_2$ layer using reactive ion etching for 4 min 5 s (Oxford Instruments, PlasmaPro NGP80, 100 W, 15 mTorr, CHF$_3$: 12 sccm, Ar: 38 sccm). In a final step, the resist was removed by sonication in acetone and isopropanol for 2 min each, followed by O$_2$ plasma ashing (PVA TePla, GIGAbatch 310M, 600 W, 5 min).

To reduce surface adhesion during template stripping, the patterned 1.5×1.5 cm$^2$ SiO$_2$/Si templates were functionalized with an ODTS self-assembled monolayer (SAM) following a recipe by Lessel et al.$^{51}$ In short, the SiO$_2$/Si template and a Teflon beaker were cleaned in a piranha solution (1:1 mixture of H$_2$SO$_4$ and H$_2$O$_2$; Caution: This solution reacts violently with solvents and other organic material) for 10 min and rinsed in DI water. The solution for SAM coating was prepared in the Teflon beaker with screw cap by first adding 150 μL of ODTS inside a N$_2$ glovebox, followed by 300 μL of CCl$_4$ and 10 mL of bicyclohexyl. Further processing steps were performed under ambient, but the Teflon beaker was kept closed whenever possible. A separate bowl of DI water was heated on a hotplate to just below the boiling temperature, and the SiO$_2$/Si templates were quickly passed over the water vapor. Immediately after the shimmering film of condensed water visually disappeared, the template was placed into ODTS solution in the Teflon beaker for 15 min. Next, to remove unbound ODTS, the template was thoroughly rinsed
with chloroform and subsequently sonicated in a chloroform bath for 2 min. To complete the process, ODTs coating, sonicating and rinsing of the SiO₂/Si chip was repeated two more times.

**Template-stripping plasmonic Ag lattices.** A schematic of the process is shown in Figure 1a in the main text. A thermal evaporator (Kurt J. Lesker, Nano36) was used for the Ag-film deposition. A nominally 25-nm-thick Ag film was evaporated at high rates (>25 Å/s) and low residual gas pressures (3×10⁻⁷ mbar) onto the plasmonic-lattice templates using Ag pellets in a tungsten dimple boat. After thin-film evaporation, Ag deposited onto the SiO₂ surface was carefully removed using adhesive tape whereas the Ag disks inside the circular holes remained. To achieve the best results the tape was gently lowered onto the template and not pressed on with a finger. Figure S1 shows a scanning electron micrograph of a removed Ag film. To facilitate imaging, here the tape was replaced by a mechanically more robust PF Gel-Film from Gel-Pak. Adhesives curable with ultraviolet (UV) light and exhibiting tunable refractive indices (Norland Products, NOA 63 and NOA 170) were then used to template-strip the Ag disks out of the etch pits. This involved adhering a glass slide onto the template using a drop of epoxy and curing it for ~1 h with a 365-nm UV lamp. The Ag-disk/epoxy/glass stack was then separated from the SiO₂/Si template by placing a razor blade between the chip and the glass slide. The SiO₂/Si template can be reused after cleaning by first sonicating it in nitric acid, then DI water, and finally chloroform, each for 5 min. After cleaning, the chip was dried using a N₂ gun.

**Integration of QD film for gain.** CdSe/CdS/ZnS core/shell quantum dots (QDs) (~4.1 nm core diameter) emitting at ~1.98 eV (625 nm) were synthesized according to a protocol described in the Supporting Information of Kress et al. The QDs were drop-casted onto template-stripped plasmonic lattices. The QD dispersions for drop casting were prepared using a 9:1 (v/v) mixture of hexane and octane and their concentration was determined by using an UV-visible spectrometer (Varian, 50 Scan using a cuvette with a 1-cm path length) to measure the optical density (OD) at the lowest-energy exciton peak. Prior to drop casting, all QD dispersions were passed through a 0.2-μm polytetrafluoroethylene (PTFE) syringe filter.
(VWR, 13 mm diameter). Finally, QD films of different thickness were prepared by drop casting 20 µL of QD dispersions with concentrations corresponding to an OD of ~2 and ~8 onto the plasmonic lattice. Slow and homogeneous drying of the QD films was ensured by covering the plasmonic lattices with a petri dish immediately after drop casting. To provide a symmetric refractive index environment and to seal the sample, the QD film was covered with the same epoxy (thickness: ~100 µm) that was previously used for template stripping. Before curing the adhesive for ~1 h with a 365-nm UV lamp, a glass coverslip was added on top to flatten and protect the capping epoxy film.

S2. Optical Measurements

Transmission measurements. The optical setup used is depicted in Figure S2. To measure momentum-resolved transmission spectra, the plasmonic lattice was mounted on an inverted microscope (Nikon, Eclipse Ti-U) and excited with a fiber-coupled broadband Xe-plasma lamp (Energetiq, EQ-99XFC). The head of a multimode fiber (core diameter: 550 µm; Thorlabs, M47L) connected to the lamp was imaged onto the plasmonic lattice using an achromatic lens (La in Figure S2 with focal length fa = 25 mm) together with a 40× air objective [Nikon, CFI S Plan Fluor ELWD; numerical aperture (NA) of 0.6]. The transmitted light was collected with a second objective of the same type (Nikon, CFI S Plan Fluor ELWD; NA of 0.6). The tube lens of the microscope (L1 in Figure S2) forms a real image of the sample on the plane labeled as “Image” in Figure S2. To project the Fourier image onto the entrance slit of the imaging spectrograph (Andor, Shamrock 303i; 150 lines/mm grating blazed at 500 nm) an achromatic lens (L2 in Figure S2) with focal length f2 = 200 mm was then used to collimate the real image from L1. A Fourier transform of the image was formed 200 mm behind the collimating lens L2. A two-lens imaging system (with achromats L3 and L4 in Figure S2 with focal lengths f3 = f4 = 200 mm) was used to project the Fourier image onto the entrance slit of the spectrograph. We switched between collecting the Fourier image and the real-space image by removing L3. Dispersion relations were obtained by spectrally dispersing the Fourier image. The dispersion relation could be probed for various in-plane wavevector
directions by physically rotating the sample. Optionally, a broadband polarizer (Thorlabs, LPVISE100-A) could be placed before the final imaging lens to record polarization-resolved data. All images were recorded using an Andor Zyla 4.2 PLUS sCMOS camera. As a reference sample for the transmission measurements of our plasmonic lattices, we recorded spectra through a blank epoxy film.

**Photoluminescence measurements.** For photoluminescence measurements, a setup similar to the one used for the transmission measurements was employed. As depicted in Figure S2, a 385-nm light-emitting diode (LED, Thorlabs, M385LP1) was used to excite the QD film through the bottom 40× air objective (Nikon, CFI S Plan Fluor ELWD; NA of 0.6). The photoluminescence signal was collected with the same objective. The excitation light was filtered from the emission signal using a 405-nm dichroic beam splitter and a 409-nm long-pass emission filter (both from AHF analysentechnik). Dispersion relations were obtained by spectrally dispersing the Fourier image using the same optics as for the transmission measurements.

**Lasing measurements.** To achieve population inversion, the LED excitation was replaced with 405-nm pulsed laser illumination (~340 fs pulse duration, 10 kHz repetition rate), generated by a collinear optical parametric amplifier (Spectra-Physics, Spirit OPA) pumped by a 1040-nm laser (Spectra-Physics, Spirit 1040-8) (see Figure S2). The collimated laser beam was directed through a gradient neutral-density filter wheel (Thorlabs, NDC-50C-2M-B) on a homebuilt motorized rotation stage to adjust the pulse power. After directing a small fraction of the laser power onto a reference power meter (Thorlabs, DET110), the remainder of the beam was defocused onto a plasmonic lattice using a defocusing lens (Lb,fb = 150 mm) together with a 40× air objective (Nikon, CFI S Plan Fluor ELWD; NA of 0.6). The emitted signal from the QDs was collected with the same objective and the laser light was filtered using a dichroic beam splitter and a 409-nm long-pass emission filter (both from AHF analysentechnik). Dispersion relations above threshold were obtained using the same optics as for the transmission/emission measurements. Power-dependent dispersion relations were measured by automatically varying the incident power using
a Matlab routine. The power was first set to the desired value by turning the neutral-density filter wheel with feedback from a power meter, and subsequently the dispersion relation was recorded with the CMOS camera.

Calibration measurements of the incident power were performed directly after the lasing measurements, by placing a second power meter on top of the objective and measuring the laser power simultaneously in the focus of the objective and at the reference power meter. Hence, we could obtain a power conversion factor between the reference power meter and the sample, which allowed us to obtain the excitation power during the lasing experiments. To calculate the pump fluence, we measured the laser-spot size by fitting a two-dimensional (2D) Gaussian function to the excitation spot and then obtained a value for the area via the extracted full-width-at-half-maximum.

S3. Lasing in Square Arrays

**Energy splitting of the lasing peak.** Figure S3 shows a magnified version of the central panel from Figure 2c in the main text. We observe two lasing peaks separated by ~2 meV. The origin of those two peaks remains unclear. We exclude that they are due to minimal differences in the pitch along the x- and y-direction (for example, due to strain induced by template-stripping) as a reason for the two peaks. Indeed, we observe the same peak splitting in rectangular lattices (see Figure 3c in the main text), where the different lattice spacings along the two orthogonal directions lead to significant energy splitting of the two corresponding resonances. We further do not anticipate that local variations in the refractive index or the pitch, which would influence the resonance condition, are the cause of the splitting. It would be surprising that such local variations are discrete, leading to two distinct lasing peaks. A broadening of the lasing peak would rather be expected.

Instead, the two peaks could result from higher-order longitudinal or transverse modes of our finite-size plasmonic array. Alternatively, minor imperfections in our plasmonic lattice could create local variations in scattering rates by the particles, effectively establishing two separate lasing cavities on the lattice with
weak cross-talk. Measuring real-space intensity patterns of the plasmonic lattice above threshold could further test these possible origins of the two peaks. Time-dependent experiments may also provide useful information. Possibly, stimulated emission in our samples occurs on a similar timescale as charge-carrier thermalization in the gain material. This could result in two bursts of laser output upon pulsed excitation separated by approximately a picosecond. The two bursts would have different photon energies because of the refractive index dynamics of the gain material, which results in changing diffraction conditions on the lattice. For each of the three options mentioned, one might expect different lasing thresholds for the different lasing peaks. We have not yet observed these experimentally, possibly because the differences are minor.

**Lasing thresholds in lattices with 380 nm pitch.** Figure S4a shows the below- and above-threshold dispersion relations of a square lattice with pitch \( a_{x,y} = 380 \text{ nm} \) and a Ag disk diameter of \( d = 70 \text{ nm} \). We observe a single lasing mode at \( \sim 1.96 \text{ eV} \) arising from the high-energy edge of the stop gap. Compared to the data shown in Figure 2c in the main text (pitch \( a_{x,y} = 390 \text{ nm} \)) the lasing energy is shifted by 30 meV consistent with eq 3 in the main text. Figure S4b shows the output intensity as a function of pump fluence. At \( \sim 0.11 \text{ mJ/cm}^2 \) the output intensity increases non-linearly, depicting typical threshold behavior for lasing. Compared to the threshold value reported in Figure 2d of the main text, the value measured for this array with smaller pitch is 0.05 mJ/cm\(^2\) lower. This difference in threshold can be explained by the better spectral overlap of the feedback-providing mode with the gain spectrum of the QDs.

**S4. Lasing in Rectangular Arrays**

**Polarization-dependent lasing emission.** Figure S5a shows the same above-threshold, polarization-resolved dispersion relations of a rectangular-lattice laser as shown in Figure 3g of the main text but plotted on a logarithmic intensity scale. While one might expect pure s- and p-polarization for the low- and high-energy lasing peaks, respectively, the logarithmic intensity scale reveals weak leakage radiation of the opposite polarization. This can be explained as a finite-size effect of the plasmonic-lattice array,
which leads to broadening of reciprocal-space features.$^{54,58}$ While the Fourier spectrum of the in-plane field profile of a bound electromagnetic mode on an infinite array consists of a series of delta peaks, these peaks are broadened for the finite-size array. The possible values of momentum provided by the plasmonic particles are also no longer delta peaks (at $i\vec{G}_1 + j\vec{G}_2$; eq 1 in the main text), but broadened. Experimentally, these broadening effects are observed in the Fourier image shown in Figure 3e of the main text. There, the lasing signal is not only emitted at $k_x = k_y = 0$, as would be expected for an infinite array, but over a small range of momentum values.

As described in Section S2, we measure the dispersion relations using the setup depicted in Figure S2. We project the Fourier image of the emission from the array on the entrance slit of the spectrograph. If the slit is aligned along $k_x$, only a small but finite range of $k_y$-values around $k_y = 0$ are transmitted. For the bound modes scattering out by coupling to $[i,j] = [\pm 1,0]$ (red circles in Figure S5b; isofrequency contours at $E_x \approx hc/n_{\text{eff}}a_x$), this selects only the outscattering near $k_x = 0$ (inset of Figure S5b). A wider range of outscattering wavevectors is selected for bound modes scattering out through coupling to $[i,j] = [0,\pm 1]$ (blue circles in Figure S5b; isofrequency contours at $E_y \approx hc/n_{\text{eff}}a_y$). Hence, a mode with frequency $\omega$ scattering out via $[i,j] = [0,\pm 1]$ appears more broadened in the $k_x$-direction than one scattering out via $[i,j] = [\pm 1,0]$. This is why the high-frequency laser mode (feedback on periodicity $a_y$) in Figure S5a is spread over a larger momentum range than the low-energy mode (feedback on periodicity $a_x$).

The finite array size affects the output polarization of the different lasing modes. The black arrows in the inset of Figure S5b indicate the orientation of the electric field relative to the isofrequency contours in momentum space. As the lasing modes are transverse-electric, the polarization direction is tangential to the contours. At the second-order Bragg condition, the $[\pm 1,0]$ modes are thus $y$-polarized at $k_x = k_y = 0$, while the $[0,\pm 1]$ modes are $x$-polarized. Hence, for a lasing mode without momentum broadening, pure output polarization would be expected, with perpendicular polarization for the two
resonances. However, if higher $k$-components scatter out (because of finite-size broadening or the finite slit width), other polarization components appear. This causes the observed leakage radiation in Figure S5a. The effect is strongest for the high-energy peak ($\{0, \pm 1\}$ modes), because a larger range of $k$-components of this peak is transmitted by the slit (see Figure S5b). The effects we discuss here may also have contributed to the observation of polarization leakage in the work of Pourjamal and coworkers.\textsuperscript{59} Indeed, they observe polarization leakage mostly for the mode broadened in momentum in the direction of the slit.
S5. Supplementary References


Figure S1. Scanning electron micrograph of the Ag film removed from the plasmonic-lattice template. The image shows the Ag film attached to a PF Gel-Film stamp after removing it from the template. The visible hole-array structure demonstrates that Ag was only removed from the top SiO$_2$ surface whereas the Ag disks remained inside the etched depressions and could later be template-stripped using epoxy.
Figure S2. The Fourier imaging setup used to obtain momentum-resolved transmission and emission spectra. The lenses $L_i$ with focal length $f_i$ (see Section S2 above for details) project the Fourier image onto the entrance slit of the spectrograph.
**Figure S3.** Angle-resolved emission maps of a Ag square lattice ($a_x = a_y = 390$ nm) at the lasing threshold. (a) Same plot as depicted in the central panel of Figure 2c in the main text. (b) Data magnified around the lasing peaks.

**Figure S4.** (a) Angle-resolved emission spectra for a square lattice ($a_x = a_y = 380$ nm) covered with a 180-nm-thick QD film. Below- and above-threshold measurements are shown on the left and right side, respectively. (b) The emission intensity from the lasing mode as a function of pump intensity, showing a threshold at 0.11 mJ/cm$^2$. 
Figure S5. (a) The same polarization-resolved measurements of the lasing emission shown in Figure 3g of the main text but plotted on a logarithmic scale. The s-polarized and p-polarized signal are shown on the left and right side, respectively. (b) Fourier image of the isofrequency contours of the high- (blue lines) and low-energy (red lines) lasing peaks. The two horizontal lines indicate the entrance slit of the spectrograph that selects a slice of the Fourier image at $k_y = 0$.

Figure S6. (a), (b) The same Fourier images as in Figures 2e and 3e of the main text, respectively. Additionally, background-corrected crosscuts along the $k_x/k_0$ and the $k_y/k_0$ axis (white dashed lines) are provided. To quantify the beam divergence, we determine the momentum range over which the intensity drops to $1/e^2$ times the peak intensity (shaded area below the cross-section curves) and report the values next to the curves. The particular intensity patterns may vary as the pump intensity changes.