

Lasing from Brillouin Zone Folding Guided Resonances

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ABSTRACT

High quality factor (Q-factor) nanophotonic cavities are critical components in applications such as lasing and non-linear optics. However, to obtain out-of-plane lasing emission and low lasing threshold, the lasing mode must fulfil the contradictory requirement of coupling to the light cone while maintaining a high Q-factor. One relatively unexplored method to design such modes consists of using a Brillouin Zone folding guided resonance (BZF-GR) as the high Q-factor mode for lasing. In such a design, guided modes are ‘folded’ into the light cone via periodic perturbations, allowing a fine control of the Q-factor throughout momentum space. In this paper, we experimentally demonstrate the use of such a BZF-GR to achieve vertical emission lasing from a nanophotonic cavity with colloidal quantum dots as a gain medium. The lowest lasing threshold under nanosecond pump is (1.88 ± 0.03) nJ per pulse. This is equivalent to (20.4 ± 0.4) kW/cm² average intensity during the pulse or (4.08 ± 0.08) kW/cm² when considering the absorbed power. This work presents a new method of designing lasing modes that may be further developed for use in low threshold nanoscale lasers.

INTRODUCTION

Lasers incorporating solution processed gain media have garnered intense attention due to their apparent ease of fabrication, and great progress in lowering the lasing threshold with improvements from both material and cavity design concepts¹⁻⁶. One convenient approach is to use a “lateral” cavity, where optical feedback is in the substrate plane, usually based on guided modes. Such designs often involve sub-wavelength patterned periodic structures, which can be broadly classified as metasurfaces, to engineer the lateral mode coupling. This allows the laser to be fabricated by depositing a modest thickness of gain material on a pre-patterned periodic structure to form the whole cavity²⁻⁸. To design a metasurface laser with out-of-plane,

or surface emission, there are several broad design considerations. A high Q-factor lasing cavity mode is necessary as it exhibits low radiative losses and a long photon lifetime, which aids the generation of optical gain and eventually lasing in the system^{9,10}. However, to obtain out-of-plane emission, it is also necessary for the mode to have a finite coupling to the light cone, which reduces the Q-factor. These opposing factors necessitate fine control of the mode radiation constant to balance external coupling, while maintaining an acceptably high mode Q-factor and hence low threshold for lasing^{11,12}. Maintaining a low lasing threshold is significantly more important when sensitive emitters such as quantum dots (QDs) are used as the gain medium as they tend to degrade more easily, and may be damaged by optical excitation before the lasing regime is reached^{2,7}.

A popular pathway to design surface emitting metasurface laser modes uses Bound States in the Continuum (BIC) as a design template^{13,14}. At the BIC point, external radiation is suppressed by cancellation of multiple radiative channels through destructive interference¹⁵. An isolated BIC at k_{BIC} , i.e. one with no other BICs nearby in parameter space, typically has a Q-factor falloff as the in-plane wavevector (k) deviates from the BIC of the form $Q \propto \frac{1}{|k - k_{\text{BIC}}|^2}$ ^{16,17}. Coupling to the light cone is expected to be from the quasi-BIC states in the vicinity of the BIC with lower Q as the BIC point itself cannot couple to the far field. This leads to a “donut” or “lobed” radiation pattern typically observed in lasing from BICs, which might be undesirable for directed emission applications such as imaging or communication^{18–20}. In a realistic device, there are additional complications. Firstly, in finite-size metasurfaces, the accessible momentum states are discretized, typically leading to a lower accessible Q-factor than at the precise BIC point^{21,22}. Secondly, fabrication limitations introduce disorder which couple the target mode with other lossy channels, also lowering the effective Q-factor²³. To counter these effects, merging- or “super”-BIC designs have been proposed and demonstrated. The idea of such designs is to position several BICs in the vicinity of a desired k-space point

to reduce the often quoted $Q \propto 1/|\mathbf{k}|^2$ falloff and scattering losses. This can be done by splitting a high topological charge BIC into separate BICs, or by tuning separate BICs together^{10,16,17,24}.

A conceptually different method to generate high Q-factor states in the light cone is Brillouin Zone Folding (BZF). An elementary unit cell with periodicity (P_0) is first defined to obtain high Q-factor guided modes that are not coupled to the light cone, and consequently, have theoretically infinite Q-factor. A weak, controlled, periodic perturbation with period $P_1 > P_0$ can be applied to extend the real space period to P_1 and reduce the BZ size. In the perturbative limit, this remaps the modes of the original structure into the new “folded” BZ, and modes now coupled to the light cone by this P_1 perturbation largely retain their original characteristics, especially their high Q-factor^{11,12,25,26}. The symmetry of the resulting supercell of period P_1 determines which (if any) modes become symmetry protected BZF-BICs or leaky guided resonances (BZF-GRs) at high symmetry points²⁷. BZF-BICs are essentially symmetry protected BICs, but with a reduced Q-factor falloff ($Q \propto 1/\alpha^2|\mathbf{k}|^2$, where α is a geometric asymmetry factor) from the BIC point. On the other hand, BZF-GRs have a broadband, relatively flat Q-factor ($Q \propto 1/\alpha^2$) with no symmetry forbidden radiation^{12,25}.

The high Q-factor attainable by this BZF design has been experimentally demonstrated by several authors. For example Wang et al. used a 1D (C_2 symmetry-breaking perturbation) design of air holes in silicon²⁵. Huang et al. also discussed the generality of a weak perturbation approach, and demonstrated a high-Q-factor silicon-on-insulator metasurface using a patterned photoresist layer on silicon, which was also demonstrated by Fang et al.^{11,28}. In plasmonic systems, de Gaay Fortman et al. have also recently demonstrated high Q factor BZF modes with a silver nanoparticle array²⁹. BZF modes may additionally be more robust to disorder than conventional BIC modes, making this design more amenable to large scale fabrication and process integration²⁵. Here we focus on engineering the fundamental Transverse Electric (TE)

BZF-GR mode in a two-dimensional (2D) metasurface to obtain vertical emission lasing with an optically pumped colloidal QD gain medium. Our lowest obtained threshold with 532 nm nanosecond pump is (1.88 ± 0.03) nJ per pulse, which corresponds to (20.4 ± 0.4) kW/cm² peak intensity, or (4.08 ± 0.08) kW/cm² when considering the absorption of the material at the pump wavelength, which compares favorably with other metasurface laser designs using similar emitters³⁰.

RESULTS

Concept and Design.

The lasing cavity consists of a TiO₂ cylinder metasurface planarized by cured Hydrogen Silsesquioxane resist (HSQ) with a film of QDs deposited on top. A quartz substrate is used with the top cladding being air. A cutaway section of an array and the corresponding periodic supercell are schematically illustrated in Fig. 1(a). The supercell is square and consists of 4 TiO₂ cylinders, arranged in a square array equidistantly from each nearest neighbor with the radius of one cylinder (r_1) different from the other three (r_2). For fixed layer thicknesses (e.g. TiO₂ = 150 nm, QD = 135 nm), tuning the radii of the TiO₂ cylinders allows the GR modes to be tuned to match the QD gain spectrum, which can be approximated by the amplified spontaneous emission (ASE) spectrum (Fig. 1(b)). The gain medium is a solution-processed film of compact-continuously-graded QDs (CdSe/Cd_xZn_{1-x}Se/ZnSe_{0.5}S_{0.5}/ZnS). The QDs are synthesized with built-in asymmetric strain, a structure which can reduce the material gain threshold by splitting the band edge degeneracy while keeping a low Auger recombination rate^{1,31,32}. By finely tuning the thicknesses of successive shells, small and highly monodisperse QDs with narrow photoluminescence (PL) of ~19 nm full width at half maximum (FWHM) in solid thin film are synthesized. Under 532 nm nanosecond stripe pump, the ASE spectrum is concentrated around the biexciton emission (<5 nm FWHM at ~643 nm). The deposited QD

films exhibit low roughness, allowing for reduced scattering losses in the metasurface modes. For enhanced stability, the solution-processed films are prepared and encapsulated in nitrogen atmosphere. Further details on the synthesis and characterization of the QDs and on the gain material characterization are contained in the experimental methods (resp. in Supporting Information sections 1 and 2).

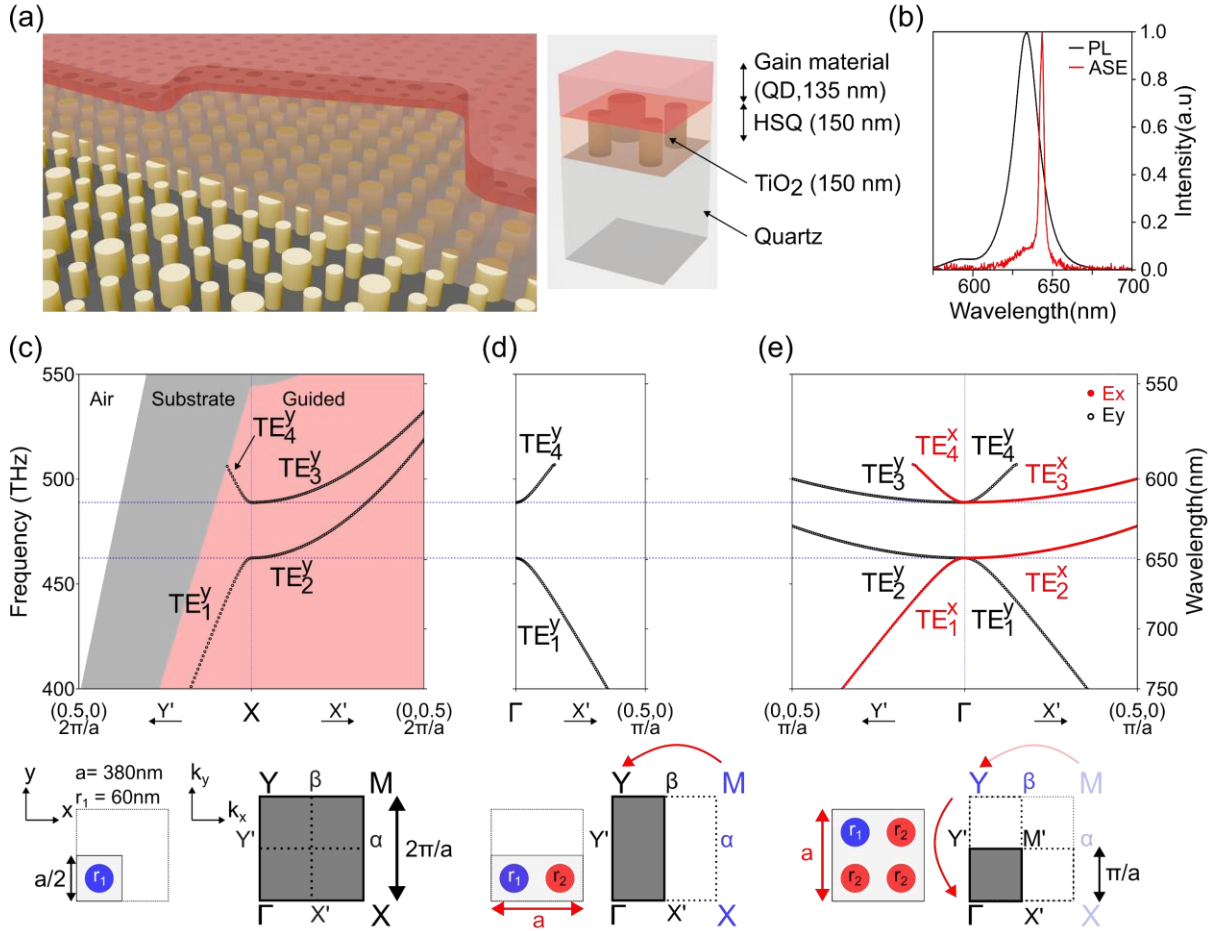


Figure 1. Brillouin-zone folding lasing cavity design concept. (a) (Left) A schematic of the lasing device composed by a QD thin film on a TiO₂ nanoantenna array planarized by HSQ on a quartz substrate. (Right) A cross-section view of the unit cell which contains 4x TiO₂ cylinders of 150 nm height and a QD film of 135 nm. (b) Normalized emission spectra of the QD gain medium under weak excitation (PL) and amplified spontaneous emission (ASE) conditions with nanosecond stripe pump. (c-e) Evolution of the calculated transverse electric (TE) band structure based on BZF from 1x1 single-cylinder cell to a 2x2 supercell when r_1 and

r_2 are considered to have an infinitesimal perturbation. **(c)** TE^y modes along $\Gamma \rightarrow X \rightarrow M$ near X of a 1×1 single-cylinder cell. For clarity only the lowest TE guided modes are shown. **(d)** Modes of a 2×1 supercell, with $P_x = 2 P_y = a$, along $\Gamma \rightarrow X$, modes $TE_{1,4}^y$ are translated from X to Γ **(e)** Modes of the 2×2 supercell along $Y' \rightarrow \Gamma \rightarrow X'$. Along $X \rightarrow X'$ 2 additional folded modes, with E_x polarization appear. Along $Y \rightarrow Y'$ the mode structure is identical, but with orthogonal polarizations. The blue line at the TE band gap serves as a guide to the eye.

From the BZF perspective, the final band structure may be considered to evolve from that of a 1×1 single-cylinder cell to a 2×2 supercell through sequential “folding” operations, for instance, where the original BZ is folded once along the x, and then y direction. Here, we limit our discussion of the band folding to the TE modes for clarity (Figs. 1(c-e)). As an example, we consider the single cylinder cell of $a/2 = 190$ nm in period and cylinder radius $r_1 = 60$ nm. Figure 1(c) shows the calculated TE modes (in black circles) along the $\Gamma \rightarrow X \rightarrow M$ direction of the original BZ near the X point. The fundamental modes near the X point are guided modes, and are E_y polarized. The modes are thus labelled TE_i^y from lowest to highest frequency. Due to symmetry, an identical band structure exists near Y point (along $\Gamma \rightarrow Y \rightarrow M$ direction) but with E_x polarized modes. Introducing an infinitesimal perturbation on the radius of every other cylinder along the x-direction the unit cell is now a 1×2 bi-cylinder cell ($1r_1, 1r_2$). P_x is doubled, so the BZ folds in half along k_x with $X \rightarrow M$ folding onto $\Gamma \rightarrow Y$. The $X \rightarrow X'$ branch is therefore folded to $\Gamma \rightarrow X'$ (Fig. 1(d)). Secondly, expanding from a 1×2 to a 2×2 supercell ($1r_1, 3r_2$), P_y is doubled resulting in a k_y -directed folding $Y \rightarrow \beta$ onto $\Gamma \rightarrow X'$. As a result, along $\Gamma \rightarrow X'$ we not only observe the $TE_{1,4}^y$ modes but also the $TE_{2,3}^x$ modes that originate from $Y \rightarrow \beta$ in the unfolded dispersion (Fig. 1(e), right panel). Symmetrically, a similar situation happens along $\Gamma \rightarrow Y'$ (Fig. 1(e), left panel). It should be noted that although all these modes are TE polarized based on their internal field, in measurements in which a linear polarizer is used, half of the modes will appear in the s- or p-polarizations. For instance, if s- and p-polarizations are defined

with the polarizer aligned to the x- or y-direction respectively, TE^x modes will appear s-polarized, and TE^y modes will appear p-polarized.

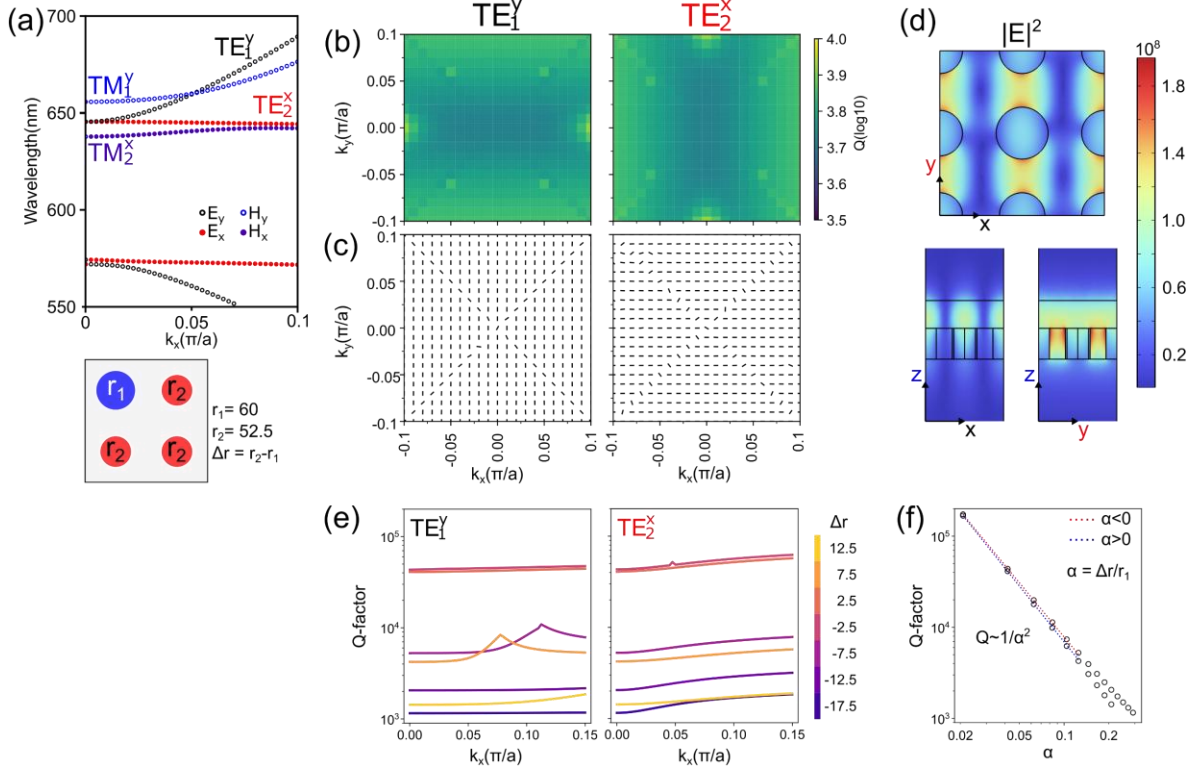


Figure 2. Mode analysis for BZF structure with a finite radius perturbation. (a) Calculated band structure for both TE and TM modes of a 2x2 supercell with $(r_1, \Delta r) = (60, -7.5)$ nm along $\Gamma \rightarrow X'$. (b) 2D k -space maps of Q-factor and (c) far field polarization maps of near Γ point of TE_1^y (left) and TE_2^x (right) for $(r_1, \Delta r) = (60, -7.5)$ nm. (d) Corresponding mode profile $|E|^2$ of TE_1^y mode at Γ point along xy (middle height of TiO_2 cylinders), xz and yz planes. (e) Simulated Q factor of TE_1^y and TE_2^x modes with $r_1 = 60$ nm and Δr varying from -17.5 nm to 12.5 nm along k_x . (f) Q-factor at Γ point scales with the asymmetry factor ($\alpha = \Delta r/r_1$) showing the $Q \propto 1/\alpha^2$ dependence. The Q-factor dependence differs slightly with the sign of α , likely due to the simplified definition of α .

Given a finite size perturbation $\Delta r = r_2 - r_1$, these BZF modes will undergo further interaction which adjusts their mode profile and wavelength. As the Q-factor of the modes is related to the

size perturbation, we index the various design combinations of cylinder radii as $(r_1, \Delta r)$ nm. Figure 2(a) shows the band structure near Γ point of $(r_1, \Delta r) = (60, -7.5)$ nm along $\Gamma \rightarrow X'$. The Transverse Magnetic (TM) H_y and H_x polarized modes $TM_{1,2}^{y,x}$ are generally present near the TE modes as well, originating from the same folding as described for TE modes. With this design, the TE_1^y and TE_2^x bands along $\Gamma \rightarrow X'$ will become BZF-GRs and are degenerate at Γ point, while the TM modes will form non-degenerate BZF-BICs. The 2nd lowest group BZF modes (TE_3^x, TE_4^y) are separated by the original photonic bandgap, sufficiently far from the QD emission spectrum, and any originally present unfolded modes are at yet shorter wavelengths. The characteristics of the TE modes are further studied to confirm their character. Figure 2(b) shows the Q-factor and Fig. 2(c) shows the far field polarization in 2D momentum space around Γ point for TE_1^y (left) and TE_2^x (right). As expected for a GR, the Q-factor is relatively constant, while the far field is linearly polarized according to the modal field polarization. In Fig. 2(d), the mode intensity profile for TE_1^y confirms the TE character with a spatial mode-gain overlap of approximately 40%. For $r_1 = 60$ nm with varying Δr , we see that the GR mode Q-factor retains similar characteristics (Fig. 2(e)) with a relatively flat Q-factor, apart from some local modulation at finite k_x likely due to mode crossings. Extracting the Q-factor for TE_1^y , at Γ point, we find that the Q-factor approximately scales as $Q \propto \alpha^{-2}$ where $\alpha = \Delta r/r_1$. There is a slight variation in the dependence with the sign of α which is likely due to our simplified definition of α . Similar characterization for the other modes is appended in Supporting Information figures S4 and S5. Overall, the BZF-GR can sustain high Q-factor with a controllably small Δr perturbation. The spatial mode overlap is $\sim 40\%$, and it should be possible to obtain low threshold vertical emission lasing from the TE BZF-GR.

Experimental results and discussions.

A series of cavities is fabricated with period $a = 380$ nm and varying $(r_1, \Delta r)$ combinations to target the TE_1 modes in the QD gain spectrum. Details of the nanofabrication are appended in the methods and Fig. S6. The low fluence angle resolved PL verifies the general accuracy of the fabrication (Fig. S7). The lowest lasing threshold is observed in an array $(60, -2.5)$ nm which is the nominal minimum asymmetry prepared in nanofabrication, and the results are presented in Fig. 3. In Fig. 3(a), the angle-resolved PL spectra for s- and p-polarizations are shown at representative pump energies. At low pumping pulse energies, the TE GR modes have a low contrast with the PL background since at low Δr the Q-factor is high. As the TE GRs are linearly polarized, and based on our collection geometry, TE_1^y appears in the s-polarization (E_y), while TE_2^x appears in the p-polarization (E_x). The TM modes are only visible at relatively high values of Δr and couple weakly to the PL, likely due to the lower spatial mode overlap (Fig. S7). As pump power increases, the mode visibility increases significantly until only a small section around $\mathbf{k} = 0$ and at ~ 643 nm is visible. A similar effect may be seen in the back focal plane (BFP) image (Fig. 3(b)). In the real space optical images, at low power there is no obvious mode coupling, while at higher power, interference fringes are apparent in the emission from the array (Fig. 3(c)).

Extracting the light in-light out (L-L) curves and full widths at half maximum (FWHM) of the emission from the PL spectrum we obtain Fig. 3(d). Lasing is confirmed by the s-shaped curve in the L-L plot, where the non-linear increase in PL intensity is concurrent with an abrupt decrease of the FWHM from ~ 9 nm to ~ 0.4 nm (limited by the spectrograph system). However, we can see that the thresholds in both polarizations are distinct, at (1.88 ± 0.03) nJ in the p-polarization and at (2.57 ± 0.13) nJ in the s-polarization based on the FWHM reducing to 50% of the initial value. Considering the lower of the two thresholds, this is approximately equivalent to (20.4 ± 0.4) kW/cm² average power. A rough estimate of absorption of the lasing

array at the pump wavelength of 532 nm is 20% (Figs. S8-9), giving the effective lasing threshold as low as of $(4.08 \pm 0.08) \text{ kW/cm}^2$.

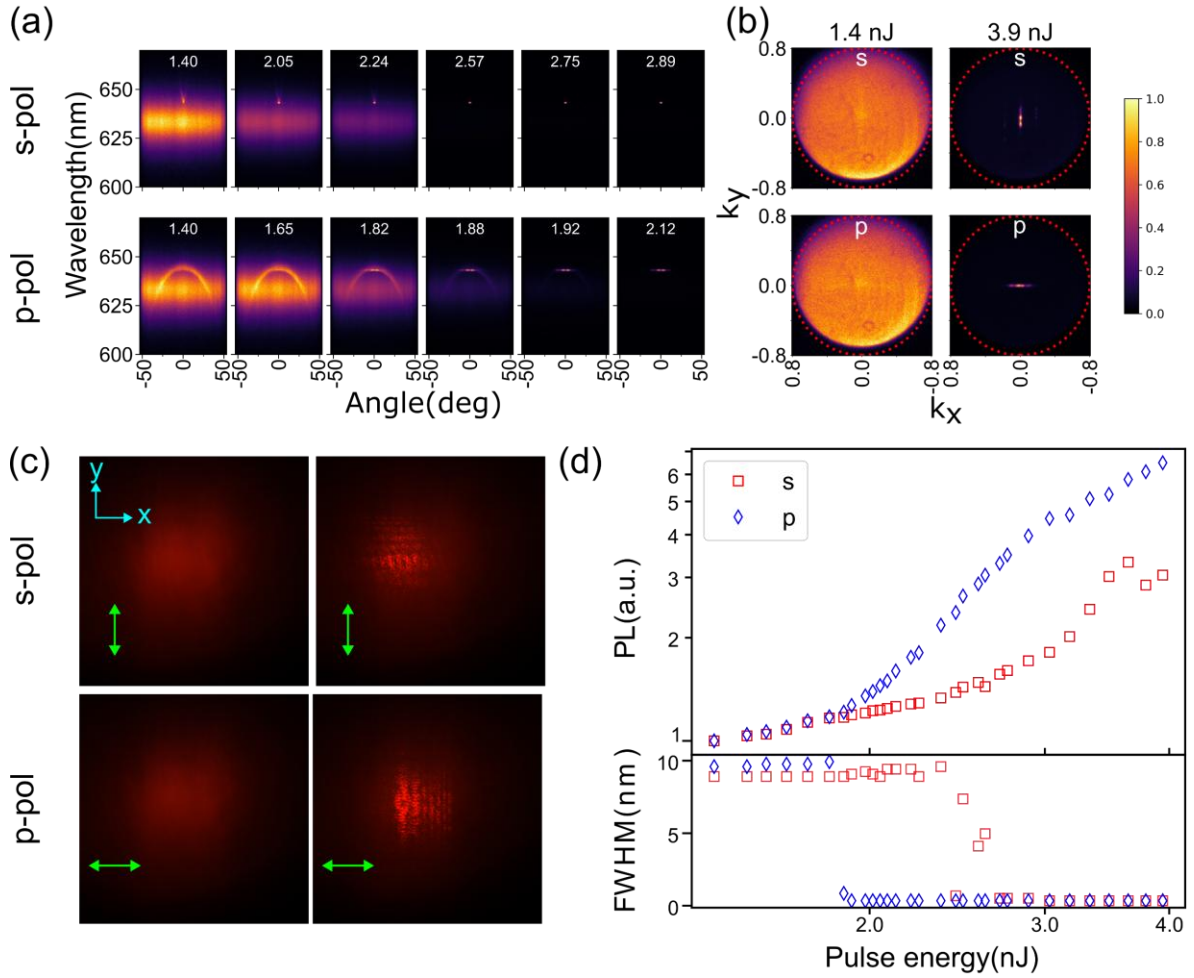


Figure 3. Lasing characteristics of the BZF-GR mode for the lowest threshold array ($r_1, \Delta r$) = (60, -2.5) nm. (a) Power dependent normalized PL spectra for s- and p-polarized emissions. Numbers in each graph indicate the incident pumping pulse energy on the sample in nJ. **(b)** Back focal plane images for s- and p-polarized emissions at low (1.4 nJ) and high (3.9 nJ) pumping pulse energies. **(c)** Optical images of the array at low (left) and high (right) pumping pulse energies under different polarizer alignments. The polarizer alignment, along with the polarization labelling is in the bottom left of each image. **(d)** Integrated PL emission (top) and FWHM (bottom) plotted against pumping pulse energy.

Many fabricated arrays (20) with varying (r_1 , Δr) also exhibit lasing behavior and we further investigate their properties (Fig. 4). The L-L curves and linewidths are extracted as described earlier, and the lasing threshold (E_{th}) is determined (Figs. S10-15). Motivated by the differing thresholds for s- and p-polarizations for (60, -2.5) nm, we characterize the lasing behavior of 2 groups of arrays ($r_1 = 50, 60$ nm) at 2 different orientations ($0^\circ, 90^\circ$, Figs. 4(a)-(d)). Generally, the thresholds in s and p-polarizations are not the same, and when lasing is observed in both polarizations at distinct thresholds, the relative threshold flips upon rotation of the sample by 90° . This indicates that the relative threshold is strongly related to physical characteristics of the array, rather than preferential alignment of the setup. However, when both polarizations lase, the lasing wavelengths remain within experimental error. A check of the lasing spectra and BFP images (Fig. S16, S17) confirms that the emission pattern is effectively the same for both polarizations, except rotated by 90 degrees in k-space. Therefore the 2 lasing polarizations are determined to originate from the 2 orthogonally polarized and aligned branches of the TE GRs (TE_1^y, TE_2^x).

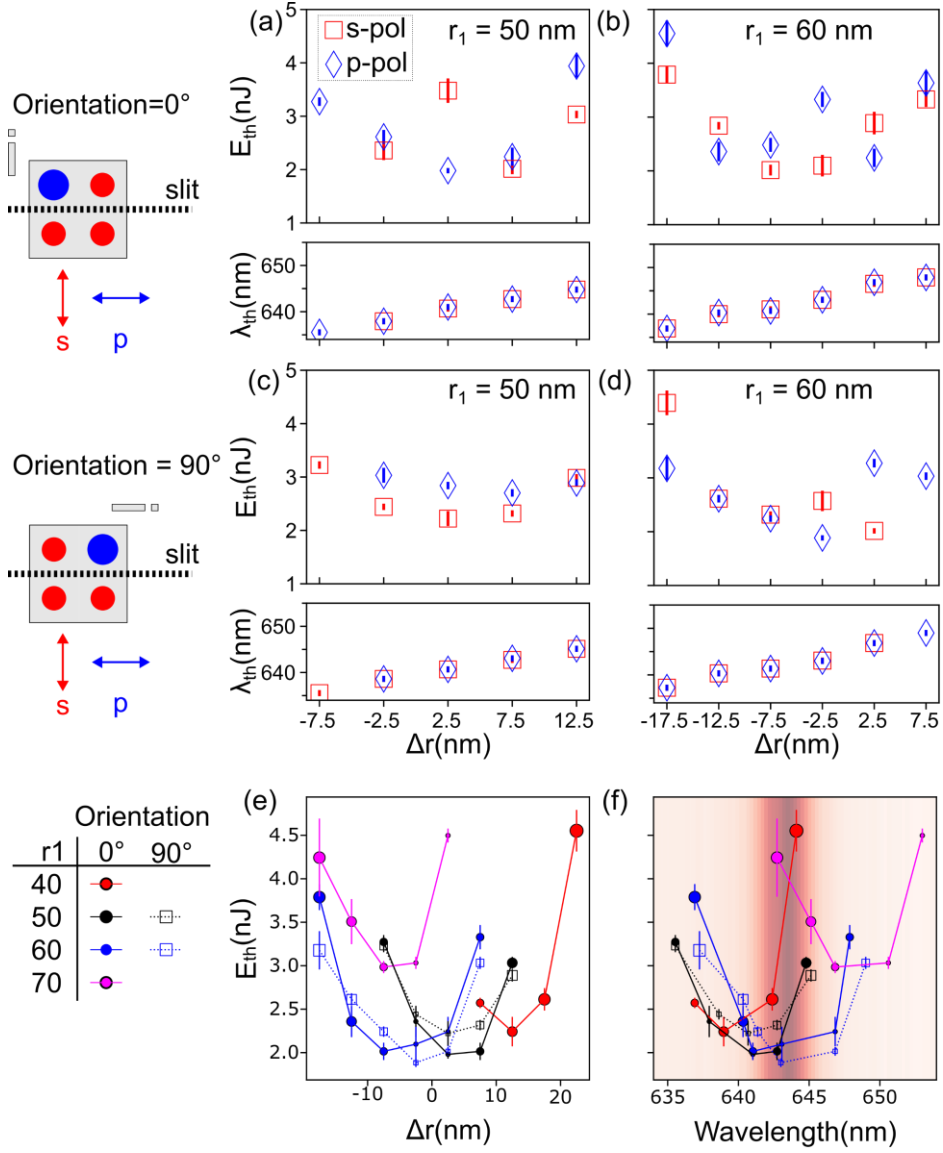


Figure 4. Systematic study of BZF-GR lasing properties. Collected results of lasing thresholds across various arrays and measurement orientations. Variation of s- and p-polarizations lasing thresholds for 0- and 90-degrees rotations for $r_1 = 50$ (a),(c) and 60 nm (b),(d). Schematics of the array rotation along with the polarizer alignment is shown to the left of the figures. Aggregated data for $r_1 = 40, 50, 60, 70$ nm and varying Δr . The lowest lasing threshold for each array is plotted against (e) Δr and (f) lasing wavelength. The ASE spectrum is superimposed in the background of (f).

As the QD emitter does not have intrinsic preferred emission direction, and we are able to observe distinct thresholds of otherwise identical modes, we still expect mode competition

between polarizations and the first mode to start lasing will suppress lasing of other modes as power is increased. Thus, the lower lasing threshold of each array is used to extract an overall trend. For threshold vs Δr (Fig. 4(e)), we also include the measurements of $r_1 = 40$ and 70 nm. We observe that the minimum overall threshold does indeed coincide with the lowest $\Delta r = 2.5$ nm. This is easily understood by the α^{-2} scaling of the Q-factor with asymmetry parameter. The lowest asymmetry (lowest Δr) array should have the highest Q-factor, and all else equal, exhibit a lower lasing threshold. However, evidently, not all groups of arrays (e.g. $r_1 = 70$ nm) exhibit lower threshold as Δr is decreased. This is also easily understood in terms of spectral overlap (Fig. 4(f)) with the gain. We see that the minimum threshold occurs when the spectral overlap with the gain is the highest and that the data points already begin to form a minimum threshold envelope centred at the ASE peak. This highlights the importance of controlling both asymmetry and mode wavelength tuning to achieve low threshold lasing, especially for emitters with narrow, concentrated gain bandwidth.

An interesting, outstanding issue is the distinct thresholds between the otherwise identical E_x and E_y polarized branches near $\mathbf{k} = 0$. It should be noted that at $\mathbf{k} = 0$, the two branches are exactly degenerate. Based on the BZF design, we can expect that the direct coupling of the k_x and k_y propagating TE modes which form the BZF-GR at $\mathbf{k} = 0$ is enhanced by approximately 4 times over a single unit cell array (e.g. with only 1 pillar in the supercell, Supporting Information section 4), while the coupling between k_x and k_y is expected to be largely unchanged^{33,34}. This may be sufficient to effectively decouple the E_x and E_y modes experimentally. For instance, defects or other non-uniformities may then separately adjust the gain threshold of each branch, leading to disparate thresholds for the otherwise degenerate modes. This effect may also be linked to the degree of spatial localization of the lasing emission evident from the optical images of the lasing arrays (Fig. S18) and is a matter for further study.

CONCLUSIONS

Overall, we have demonstrated low threshold vertical emission lasing using BZF-GR modes. Systematic studies of the lasing behavior as a function of asymmetry factor Δr and mode-gain overlap confirm that the lasing threshold can be controlled by adjusting the array asymmetry Δr and tuning of the mode-wavelength position relative to the gain spectrum. We expect that if a sufficiently small but controlled perturbation Δr is introduced, we can maximize the Q-factor of the BZF-GRs and maintain finite coupling and radiation into the vertical $\mathbf{k} = 0$ direction. Further work may employ more complex mode engineering to optimize the emission, for example to focus the laser output into a controlled divergence beam.

In this design, we also have two nominally identical, but orthogonally polarized BZF-GR modes lasing at different thresholds. With better control or dynamic tuning of the array, this may enable single mode, switchable polarization lasing. From the general design perspective, a better understanding of the system is still required to maximize performance. For instance, given the large number of methods available to generate a BZ folding perturbation, evaluating the effect of specific perturbations on the effective quality factor and gain threshold(s) of the target modes will enable more targeted design. Mode competition and escape channels will eventually need to be carefully considered to control the useful outcoupling of lasing emission from the GR, versus the lasing threshold. With careful design, this BZF system may provide an alternative approach to achieving low threshold vertical emission lasers suitable for integration with sensitive gain materials such as colloidal QDs.

METHODS

Optical characterization of BZF laser: Optical characterization is performed on a customized inverted microscope (Nikon Ti-U). A schematic of the setup is provided in Supporting

Information Figure S1. For reflectance and transmission measurements, a diffuse halogen lamp is used as the excitation source. For Photoluminescence and lasing measurements, a 532 nm ns-pulsed laser.

Nanofabrication of TiO₂ metasurfaces: The basic structure is TiO₂ pillars made by reactive-ion-etching with a Cr hardmask. Quartz substrates are prepared (JGS2, latech) by cleaning with sonication in Acetone and IPA, before O₂ plasma treatment. 150 nm TiO₂ (Oxford Optofab3000) and 30 nm Cr (Angstrom Evovac) are deposited onto the substrate. A surface treatment of Surpass 3000 (Dishcem) is spincoated at 4000 rpm for 45 s, followed by DI water at 4000 rpm for 45 s. The substrate is baked at 180 °C for 2 minutes to dehydrate it. 6 wt% HSQ in MIBK (EM-resist or Dischem) is spin coated at 4000 rpm for 45 s, then annealed at 120 °C (2 minutes) / 180 °C (2 minutes). Electron beam lithography is performed by a Elionix ELS-7000 at 100 kV acceleration voltage, 500 pA beam current. The dosage is adjusted for the arrays to remain within expected size tolerance. The HSQ pattern is developed in salty developer (4 wt% NaOH, 1 wt% NaCl, Aqueous) for 4 minutes before rinsing thoroughly with DI water. The HSQ pattern is transferred to the Cr hardmask by reactive ion etching (Oxford OIPT Plasmalab) and the TiO₂ pillars are then etched through the hardmask by reactive ion etching (SPTS APS or Oxford Plasmalab 100 Cobra). The hardmask is removed by wet etching in Cr-etchant (Sigma-Aldrich). After sample qualification, an identical cleaning procedure to the first step is performed. Concentrated HSQ, FOX-22 (Dow-corning) is spin coated at 1000 rpm for 60 s. Annealing is then performed at 300 °C for ~1 hour with a ramp up rate from <150 °C at 450 °C/hour. The sample is allowed to cool to room temperature on the hot plate. The FOX-22 is then etched down to approximately the same height as the TiO₂ pillars. Other various tools are used to calibrate the process such as SEM (Hitachi SU8 2200), reflectance (Filmetrics), Surface profiler (KLA Tencor), Variable angle spectroscopic Ellipsometry (J A Woolam).

Synthesis of compact continuously graded QDs (ccg-QDs): Materials: Cadmium acetate dihydrate ($\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$, >98%), cadmium oxide (CdO , 99.9%), zinc acetate dihydrate ($\text{Zn}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$, 99.99%, trace metal basis), selenium (Se , 99.99%, trace metals basis), sulfur (S , 99.998%, trace metals basis), trioctylphosphine (TOP, 97%), myristic acid (MA, 98%), oleic acid (OA, 90%), 1-octadecene (ODE, technical-grade), were purchased from Sigma-Aldrich. Toluene, chloroform, and ethanol were purchased from Merck Millipore and used without any further purification.

Precursor preparation: *Preparation of Se-TOP (2 M) precursor*: 2 M Se-TOP solution was prepared and stored inside the glove box, by fully dissolving the 789.6 mg (10 mmol) selenium in 5 mL TOP. *Preparation of S-TOP (2 M) precursor*: 2 M S-TOP solution was prepared and stored inside the glove box, by fully dissolving the 320 mg (10 mmol) selenium in 5 mL TOP. *Preparation of Cd-oleate ($\text{Cd}(\text{OA})_2$) (0.5 M) precursor*: In a 50 mL three neck round bottom flask, 2.67 g (10 mmol) $\text{Cd}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ was dissolved in 10 mL OA and 10 mL ODE by degassing at 120 °C under vacuum for 1 h. The final mixture is finally transferred in a sealed bottle and stored in a glove box or further uses. *Preparation of Zn-oleate ($\text{Zn}(\text{OA})_2$) (0.5 M) precursor*: The Zn-oleate solution was freshly prepared before the synthesis. In a 50 mL three-neck flask, 1.83 g (10 mmol) $\text{Zn}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ was dissolved in 10 mL OA, and 10 mL ODE by degassing at 130 °C under vacuum for 1 h and then kept at 120 °C under nitrogen gas for further use. *Shell growth solutions*: The synthesis of $\text{CdSe}/\text{Cd}_x\text{Zn}_{1-x}\text{Se}/\text{ZnSe}_{0.5}\text{S}_{0.5}/\text{ZnS}$ (ccg-QDs) was made using the four precursor solutions (A, B, C, and D) which were prepared inside the glove box. Solution A was prepared by mixing the 0.5 mL of $\text{Cd}(\text{OA})_2$ (0.5 M), 0.125 mL of Se-TOP (2 M), and 0.375 mL ODE. Solution B was prepared by mixing the 1.25 mL of $\text{Cd}(\text{OA})_2$ (0.5 M), 1.25 mL of Se-TOP (2 M), and 2.5 mL ODE. Solution C was prepared by mixing 0.5 mL of Se-TOP (2 M) and 0.5 mL of S-TOP (2 M) in 1 mL ODE. Solution D was prepared by mixing 0.5 mL of S-TOP (2 M) in 0.5 mL ODE.

Synthesis of the QDs: In a typical synthesis, 12.8 mg CdO (0.1 mmol), 69 mg myristic acid (0.3 mmol), 0.1 mL OA (0.3 mmol), and 6 mL ODE are loaded in a 100 mL three-neck round flask and degassed at 120 °C for 30 minutes before switching to nitrogen atmosphere. At 310 °C, 0.1 mL Se-TOP (2 M) is swiftly injected into the reaction mixture. After 30 s, 1 mL of TOP is added dropwise. After 2 min, 1 mL of Solution-A was added continuously into the reaction flask at a rate of 5 mL/h for 12 min. The solution is then annealed for another 1 min 30 s. For the first coating of graded $\text{Cd}_x\text{Zn}_{1-x}\text{Se}$ shell, 2 mL of $\text{Zn}(\text{OA})_2$ (0.5 M) are swiftly injected into the reaction flask at 300 °C followed by the dropwise injection of 5 mL of Solution-B at a rate of 5 mL/h for 60 minutes. During this injection, 2, 4 and 2 mL of the $\text{Zn}(\text{OA})_2$ (0.5 M) mixture were swiftly added at 12, 35.5, and 49.5 min marks respectively. For the second coating of $\text{ZnSe}_{0.5}\text{S}_{0.5}$ shell, 1.5 mL of Solution-C was continuously introduced into the reaction mixture at 300 °C with a rate of 2 mL/h for 45 min. During this injection, 1 mL of the $\text{Zn}(\text{OA})_2$ (0.5 M) mixture was swiftly added at 15, 30, and 45 min. For the last coating of ZnS shell, 0.25 mL of Solution-D was continuously injected into the reaction flask at 300°C with a rate of 1 mL/h for 15 min. During this injection, 1 mL of the $\text{Zn}(\text{OA})_2$ (0.5 M) mixture was swiftly added at 7 min. The mixture is then cooled down to room temperature, and 18 mL of chloroform were added at 70 °C. To purify the as-synthesized ccg-QDs, ethanol was added to precipitate the QDs and the mixture is centrifuged at 7000 rpm for 5 min. After centrifugation, the supernatant was discarded, and the precipitate redispersed in 10 mL toluene and stored in a glovebox for further use.

Characterization of ccg-QDs: TEM: For ccg-QDs imaging, several drops of diluted ccg-QDs solution were drop-casted on top a copper grid covered with an amorphous carbon film. Image acquisition was conducted using a JEOL TEM 2200F transmission electron microscope operated at 200 kV. AFM: QD film thickness and roughness were measured with AFM by

using the scan-assist mode by a Bruker Dimension Icon scanning probe microscope (Bruker 18 Co., Germany). The results are provided in Supporting Information Figure S2.

Optical characterization: Absorbance measurements of the ccg-QDs were recorded using a Shimadzu UV and PL spectrophotometers by using diluted solution of colloidal ccg-QDs. PL-QY measurement setup was equipped with an Ocean Optics S4000 spectrometer, and an integrating sphere was used. Colloidal ccg-QDs were excited at a wavelength of 405 nm (Cobolt Laser). For femtosecond pumped Amplified Spontaneous Emission measurements, a 400 nm pump laser was obtained by frequency doubling a femtosecond Ti:sapphire 800 nm source (Coherent Astrella F) using an OPA (Coherent Topas prime). Then, the emission was collected using a fiber-coupled ANDOR spectrometer (monochromator: ANDOR Shamrock 303i, CCD: ANDOR iDus 401). The emission spectra were collected perpendicular to the laser pump. For nanosecond pump Amplified Spontaneous Emission measurements the laser source is a 532 nm ns-pulsed laser (Crylas FDS532, ~1 ns pulse width). The laser is focused onto the sample through a cylindrical lens into a stripe of ~0.1 mm². Emission spectra is collected from the edge of the sample with a fiber-coupled spectrometer (Ocean Optics HR4000).

ccg-QD Film deposition: 200 μ L of the QD stock solution is precipitated by centrifugation (6000 rpm for 2 min) using 1:1 EtOH ratio. The precipitates were redispersed in 100 μ L of toluene. Any remaining aggregates were crashed out by centrifugation (8000 rpm for 1 min). The patterned substrates are cleaned using O₂ plasma (100 W for 10 min). Patterned substrates and QD solution are transferred to a nitrogen filled glovebox box, and 60 μ L of the solution is spin-coated on top of the pattern (2000 rpm, 1 min). The resulting film is encapsulated using a carved quartz cap and sealed using epoxy glue. The results are provided in Supporting Information Figure S3.

Numerical Simulations: The band structures, Q-factor, and far field polarization maps were simulated using finite element method (COMSOL Multiphysics version 6.2). Details of the

model parameters and generation of far field polarization maps are discussed in detail in the Supporting Information, Section 3.

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M.R.C. and L.D. conceived the idea, optical characterization, and data analysis; W.W., L.D., and M.R.C. conducted numerical simulation; M.R.C., L.D., and F.T. conducted nanofabrication; X.L. did material synthesis and growth and film encapsulation study; X.L. and C.D. did thin film preparation and encapsulation; S.A., E.G.D. conducted material and film characterization; M.C. drafted the initial manuscript with input from C.D. regarding the material part; L.D., M.R.C., and R.P.D. participated in discussion; M.C., L.D., H.V.D., and

A.I.K. revised and finalized manuscript with inputs from all authors; L.D., H.V.D., and A.I.K. supervised the whole project.

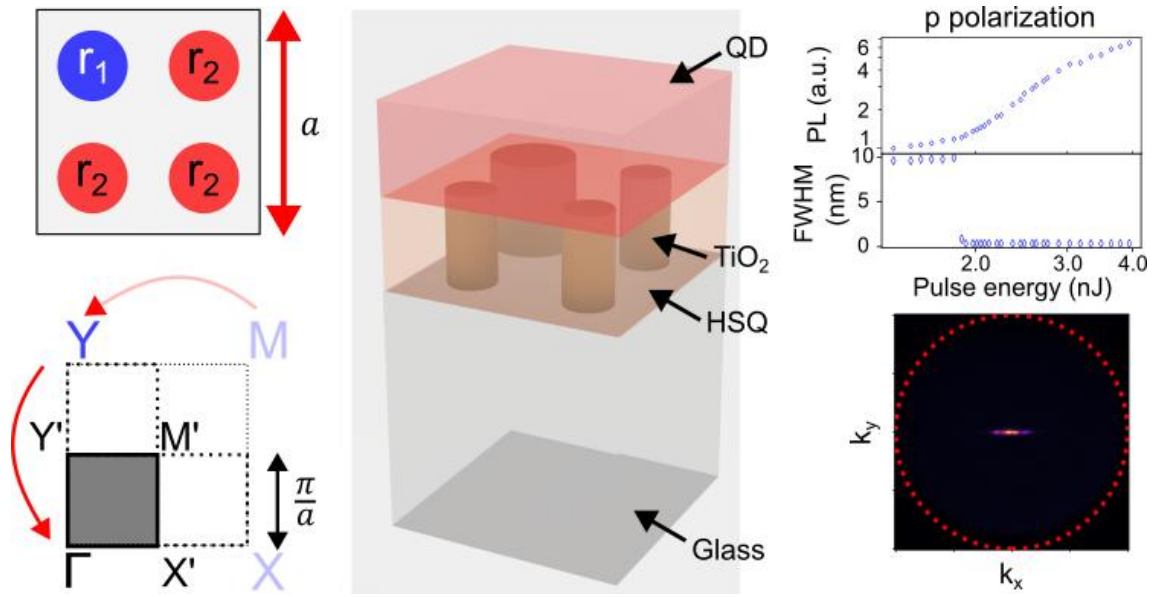
Notes

The authors declare no competing financial interest.

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TOC



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