Lasing Action from Quasi-Propagating Modes

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Band edges at the high symmetry points in reciprocal space of periodic structures hold special interest in materials engineering for their high density of states. In optical metamaterials, standing waves found at these points have facilitated lasing, bound-states-in-the-continuum, and Bose–Einstein condensation. However, because high symmetry points by definition are localized, properties associated with them are limited to specific energies and wavevectors. Conversely, quasi-propagating modes along the high symmetry directions are predicted to enable similar phenomena over a continuum of energies and wavevectors. Here, quasi-propagating modes in 2D nanoparticle lattices are shown to support lasing action over a continuous range of wavelengths and symmetry-determined directions from a single device. Using lead halide perovskite nanocrystal films as gain materials, lasing is achieved from waveguide-surface lattice resonance (W-SLR) modes that can be decomposed into propagating waves along high symmetry directions, and standing waves in the orthogonal direction that provide optical feedback. The characteristics of the lasing beams are analyzed using an analytical 3D model that describes diffracted light in 2D lattices. Demonstrations of lasing across different wavelengths and lattice designs highlight how quasi-propagating modes offer possibilities to engineer chromatic multibeam emission important in hyperspectral 3D sensing, high-bandwidth Li-Fi communication, and laser projection displays.

1. Introduction

2D photonic crystals have characteristics that are extensions of their 1D counterparts. 1D distributed feedback cavities support single-mode lasing with linearly polarized emission and longitudinal beam profiles, and because of their increased rotational symmetry, 2D cavities enable multimode lasing and vortex polarization, and annular-shaped beams. Most lasing work on 2D photonic crystals exploits band edges at high symmetry points (e.g., Γ, X, and M points of a square lattice) in reciprocal space for optical feedback. Since standing waves at these points are biaxially confined, solutions to their wave equation are critically constrained, which limits lasing action to discrete wavelengths and directions. However, 2D photonic crystals can also be considered as lines of 1D arrays, where in-plane scattered waves are decomposed along two orthogonal directions. In this picture, quasi-propagating photonic modes are slow traveling waves and can be interpreted as uniaxially confined standing waves that propagate along high-symmetry directions (e.g., Γ–M, Γ–X, and M–X). The additional degree of freedom from propagation enables the band edge states to span a continuum of energies and wavevectors. Although quasi-propagating modes are predicted to support optical feedback, lasing action via 2D cavities remains primarily focused on that from high symmetry points.

Strongly scattering 2D plasmonic nanoparticle (NP) lattices that can trap light in-plane support hybrid photonic-plasmonic modes known as surface lattice resonances (SLRs). Feedback from SLRs has enabled nanoscale lasing from NP lattice cavities integrated with index-matched emitter gain materials such as organic dyes in solvents and upconversion NP thin films. NP lattices integrated with high-refractive-index emissive materials such as colloidal quantum dot films have also demonstrated lasing from transverse electric (TE) and transverse magnetic (TM) waveguide-hybridized SLRs. Because of the mode structure of the waveguide component, W-SLRs can excite large volumes of active material for lasing. For either SLR or W-SLR modes, however, feedback for lasing is from biaxially confined standing waves since their losses are lower than quasi-propagating modes. Lasing from quasi-propagating modes should be possible; we hypothesize that they have been elusive due to insufficient gain coefficients and offer higher gain, their syntheses are challenging. In contrast, lead halide perovskite nanocrystals (NCs) can be readily
synthesized at low temperatures[34] and at large scales.[35] Importantly, their high modal gain of $\approx 450 \text{ cm}^{-1}$[36–39] has facilitated amplified spontaneous emission in solution, multiphoton upconversion stimulated emission, and ultralow threshold lasing.[40–43] Moreover, perovskite NCs have narrow gain bandwidths that can be continuously tuned throughout the visible range by post-synthesis halide exchange.[44] These optical characteristics enable the testing of quasi-propagating modes for lasing at specific energies and across different energy ranges.

Here we show that quasi-propagating photonic modes can provide feedback for lasing. Using aluminum (Al) NP lattices as optical cavities and lead halide perovskite NCs as gain, we demonstrate lasing action over a continuous range of wavelengths and directions from a single device. Quasi-propagating mode characteristics were predicted analytically using a geometric light-cone model that describes diffracted light in 2D lattices. Finite-difference time-domain (FDTD) simulations confirmed the uniaxial optical confinement of quasi-propagating modes, with a standing wave in one direction and a propagating wave in the orthogonal direction. Finally, we demonstrate the generality of quasi-propagating photonic modes for lasing action through various combinations of plasmonic lattices and perovskite NCs emitting at different wavelengths with expected emission beam patterns.

2. Results and Discussion

Figure 1a depicts a scheme of a quasi-propagating laser device. CsPbBr$_3$ NCs synthesized by previously-reported methods[34] ($\lambda_{\text{PL}} = 515$ nm, Figure S1, Supporting Information) were spin-coated as a 250 nm-thick film on 2D Al NP square lattices (periodicity $a_0 = 300$ nm, diameter $d = 60$ nm, height $h = 60$ nm) fabricated by SANE and PEEL processes[45] (Figure S2, Supporting Information). Al was selected as the plasmonic material because of its high stability[46] and low losses at both emission and pump wavelengths.[47] Because the refractive index ($n$) of the gain layer is higher ($n_{\text{NC}} \approx 1.75$ @ $\lambda_{\text{PL}}$, Figure S3, Supporting Information) than that of the quartz substrate ($n_{\text{sub}} \approx 1.45$), the optical modes are classified as W-SLRs.[21–23] When devices are optically pumped at 400 nm ($<100$ fs, 1 kHz, fluence $\approx 100 \mu\text{J cm}^{-2}$), twelve beams are emitted on each side of a device; these are symmetrically distributed about the four-fold rotational axis normal to the lattice plane. The beams can be categorized by their polar angles ($\theta$) normal to the plane and their azimuthal angles ($\phi$) in-plane. Four beams were proximal to normal at $\theta \approx 13 \pm 0.5^\circ$ and $\phi \approx 45, 135, 225, \text{ and } 315 \pm 0.5^\circ$ (Figure 1b, orange dashed box), while the other eight were distal at $\theta \approx 80 \pm 0.5^\circ$ and $\phi \approx 27, 63, 117, 153, 207, 243, 297, \text{ and } 333 \pm 0.5^\circ$ (Figure 1b, green dashed box) (Methods, Figure S4, Supporting Information). Emission of the twelve beams spans a hemisphere, but we could only capture eight simultaneously on a curved white screen (Figure 1b, inset). All of the proximal and distal beams are distinct from those observed from lasing at the high symmetry points of 2D square lattices (one beam from the $\Gamma$ point and four first-order beams from each $\Delta, X, \text{ and } M$ point based on symmetry).[7,22,48]

Spectral properties of both proximal and distal beams indicate a narrow peak centered at $\approx 526$ nm with full width at half

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**Figure 1.** Lasing from quasi-propagating modes in plasmonic lattices. a) Scheme of device where CsPbBr$_3$ NC films are integrated with Al NP lattices of periodicity $a_0 = 300$ nm and emit 12 lasing beams. b) Optical microscopy image of the lasing beams from the top when the device is pumped at 400 nm above threshold. The black square excludes the pump beam. (inset) Scheme of side view for imaging the beams. c) Emission spectrum of a proximal laser beam emitting at $\theta = 13^\circ$. (inset) Input–output curves on a log–log scale; the lasing threshold is 21.9 $\mu\text{J cm}^{-2}$. d) Angle-resolved emission measurements for a proximal beam along the axis of elongation ($\phi = 45^\circ$) reveals that the lasing wavelength at each position along the beam varies as a function of $\theta$. (inset) Proximal beam outlined in dashed orange box in (b) imaged by Fourier microscopy.
maximum (FWHM) of $<1.2$ nm (Figure 1c; Figure S5, Supporting Information), similar to previous lasing characteristics via W-SLR modes.\[21,23,49\] Both sets of beams had similar thresholds despite their different emission angles; pump fluence versus output intensity curves showed a linear-to-superlinear transition which is accompanied by a sharp decrease in emission linewidth (Figure S6, Supporting Information) at threshold ($P_{th}$) $≈ 22 \, \mu J \, \text{cm}^{-2}$. Because individual CsPbBr$_3$ NCs have degenerate transition dipole moments along their three orthogonal crystal directions,\[50\] films of randomly packed NCs emit unpolarized light isotropically and independent of excitation polarization.\[50,53\] The emitted light can therefore excite $W_{TE}$-SLR and $W_{TM}$-SLR modes along both directions of the 2D lattice\[22\] that can interfere to form quasi-propagating modes. Unlike lasing via biaxially-confined modes that exhibit vortex polarization (e.g., at the $\Gamma$ and $X$ points),\[5,22,23,52\] both distal and proximal beams show linear polarization that is consistent with uniaxial confinement;\[1,2\] the proximal beams are s-polarized, and the distal beams p-polarized (Figure S7, Supporting Information). For all beams, their highly directional emission, narrow emission spectra, and threshold behavior indicate lasing action that was stable for several minutes under sustained excitation at $1.5P_{th}$ (Figure S8, Supporting Information).

A Fourier-plane image of an objective lens (numerical aperture NA = 0.3, Figure S9, Supporting Information) indicates that the proximal beams at $\theta = 13^\circ$ are elongated with an angular spread of $\Delta \theta = 3.3 \pm 0.4^\circ$ (Figure 1d, inset). Angle-resolved spectra along the length of the proximal beam show that as $\theta$ increases along the axis of elongation, the lasing wavelength continuously shifts to lower energies with a range of $\lambda = 522.1-528.6$ nm ($\Delta \lambda = 6.5$ nm, $27$ meV); a Gaussian intensity distribution is centered about $\theta = 13^\circ$. The widths of the lasing peaks at each selected wavelength were similarly narrow (FWHM 1.0–1.4 nm) and spanned the optical gain profile of multie excitonic emission in CsPbBr$_3$ NC films.\[36,53\] These results indicate that the entire gain bandwidth contributes to lasing emission across a range of wavelengths, which is in contrast to lasing from a biaxially confined mode that occurs at a single wavelength within the gain profile. Similar to the proximal beams, mapping the pattern of a distal beam revealed a Gaussian intensity distribution with $\Delta \theta = 4^\circ$; lasing spectra at locations along the elongation shifted toward decreasing energies as $\theta$ increased (Figure S10, Supporting Information).

To interpret the locations and energies of the two sets of lasing beams, we used a 3D model of reciprocal space in the empty-lattice approximation\[10,14\] to describe optical feedback in 2D lattices (Supporting Information). We used the empty-lattice approximation as a qualitative and intuitive model to obtain information about the in-plane scattered light for different energies and lattices. The free-space photon dispersion can be represented as a mode cone with radius $|k_{mode}| = E_{n_{eff}}/c$ where $k_{mode}$ is the in-plane wavevector, $E$ the energy of the scattered wave, and $n_{eff}$ the effective refractive index. In 2D lattices, the in-plane momentum is represented as periodic cones centered at each diffraction order (Figure 2a, blue); intersections between cones result in interference and the formation of band-edge states that support optical feedback. Because the radius of a mode cone increases linearly with the energy of the scattered waves, an intersection occurs over a continuum of energies and wavevectors. Although W-SLRs can support multiple TE and TM waveguide modes of different refractive index $n_{eff}$,\[21–23\] we focus on a single mode with $n_{eff} = 1.60$ for clarity. For either polarization, however, since the radiative out-coupling of W-SLRs is from diffractive scattering, their emission can be modeled with the empty lattice approximation.

Figure 2b depicts a 2D projection of the cones that overlaps the finite gain bandwidth of CsPbBr$_3$. Since only modes that emit to the far-field can be measured, we only considered those intersections that were within the light cone in air (indicated
in red). We denote cone intersections at single energy by green circular dots, which when integrated over the finite gain bandwidth, are elliptically shaped. The four intersections close to the Γ-point indicate optical feedback for the proximal beams and the eight intersections near the border of the light cone for the distal beams. At any point along an intersection, two interfering optical modes with wavevector $k_{\text{mode}}$ (Figure 2b, black solid arrows) form a quasi-propagating mode characterized by i) a propagating wave along a high symmetry direction (Figure 2b, red arrows) and ii) a standing wave in the orthogonal direction that provides optical feedback (Figure 2b, purple dashed arrows). Quasi-propagating modes associated with the proximal beams travel along Γ–M, while those with the distal beams are along M–X. In contrast, biaxially-confined waves at high symmetry points do not propagate along any axis and only occur for specific, discrete values of $k_{\text{mode}}$ (Figure S11, Supporting Information).

To model the far-field distribution of the lasing beams, we converted the cone intersections in reciprocal space (Equations S5 and S6, Supporting Information) into spherical polar coordinates in real space (Figure 2c). The spatial distribution of the beams agrees with experiment (Figure 1b), with the proximal beams at $\theta = 13.0^\circ$ and $\phi = 45, 135, 225, \text{and} 315^\circ$ and the distal beams at $\theta = 76.2^\circ$ and $\phi = 25, 65, 115, 155, 245, 295, \text{and} 335^\circ$. The calculated beam spots are also elongated according to the gain bandwidth ($\lambda = 522.1$–$528.6 \text{ nm}$), with the emission wavelengths increasing as $\theta$ increases along a spread of $\Delta \theta = 2.0^\circ$ for the proximal beams and $\Delta \theta = 5.3^\circ$ for the distal beams (Figure 2c, inset). The higher $\Delta \theta$ for the distal beams is from the sinusoidal relationship when intersections are converted from reciprocal space into real-space polar coordinates.\(^{[54]}\)

To characterize the lasing mechanism from quasi-propagating modes, we examined the s-polarized optical band structure along the Γ–M direction (i.e., $\phi = 45^\circ$, along a pair of proximal beams). Because s-polarized light is neither orthogonal nor aligned with a 2D square lattice axis,\(^{[22]}\) the image shows two sets of bands that can be assigned as W-SLR modes (Figure 3a). Since TE modes oscillate in-plane with the high-index perovskite NC film,\(^{[55]}\) the TE-polarized modes have a higher $n_{\text{eff}}$ compared to TM waveguide modes\(^{[21,23,56]}\); hence, $W_{\text{TE}}$-SLR bands are lower in energy than $W_{\text{TM}}$-SLR bands. The narrower linewidths of the $W_{\text{TM}}$-SLRs indicate their higher quality factors ($Q$) compared to $W_{\text{TE}}$-SLRs.\(^{[24]}\) The $W_{\text{TE/TM}}$-SLR bands represent $(-1,0)/(0,-1)$ and $(0,1)/(1,0)$ diffraction orders and are obscured at energies > 2.4 eV because of strong absorption by the CsPbBr\(_3\) NCs. Angle-resolved photoluminescence measurements with pump fluences below $P_{\text{th}}$ show directional emission along the W-SLRs because of scattering by the lattice as well as a broad, non-dispersive band at $\approx 2.4$ eV ($\lambda_{\text{PL}} = 515 \text{ nm}$) from spontaneous emission (Figure 3b).

When the device is pumped above $P_{\text{th}}$, the $W_{\text{TM}}$-SLR modes that overlap the gain exhibit bright emission at $|k_y| = 2.95 \mu\text{m}^{-1}$ (Figure 3c), corresponding to proximal lasing beams at $\theta = \pm 13.0^\circ$. Because stimulated emission in CsPbBr\(_3\) NCs is from recombination of biexcitons with attractive interactions,\(^{[57]}\) their optical gain bandwidths are narrower and at energies lower than the PL bandwidth.\(^{[36,58]}\) Lasing emission from $W_{\text{TM}}$-SLR modes has higher intensity and narrower FWHM ($\leq 1.4 \text{ nm}$) than enhanced photoluminescence from $W_{\text{TE}}$-SLR modes with an FWHM ($> 2.8 \text{ nm}$) (Figure S12, Supporting Information). The difference between emission characteristics can be attributed to larger $Q$ and larger mode volumes in $W_{\text{TM}}$-SLRs that can excite higher proportions of NC via waveguide effects, compared to $W_{\text{TE}}$-SLRs with lower $Q$ that mostly excite NCs within the electromagnetic hotspots of the Al NPs (Figure S13, Supporting Information). Notably, lasing emission is observed over the entire range of energies bound by the 27 meV gain bandwidth along degenerate $(-1,0)/(0,-1)$ and $(0,1)/(1,0)$ $W_{\text{TM}}$-SLR bands that propagate along Γ–M. To confirm the standing wave characteristics in the direction orthogonal to Γ–M, we simulated the optical band structure at $\theta = 13^\circ$ along $\phi = 35–55^\circ$ (Figure S14, Supporting Information). Here, a minimum in the parabolic $W_{\text{TM}}$-SLRs band at $\phi = 45^\circ$ represents an upper band edge that supports standing waves. Other details of the quasi-propagating modes are validated by the near-field phase maps that show propagation along Γ–M and stationary character in the orthogonal direction (Figure S15, Supporting Information).

We confirmed the generality of quasi-propagating lasing across a range of energies, diffraction orders, and lattice symmetries (Figure 4). To access quasi-propagating modes at higher energies, we took advantage of the post-synthetically tunable bandgaps in perovskite NCs.\(^{[54]}\) By mixing solutions of CsPbBr\(_3\) NCs and CsPb(Cl\(_0.65\)/Br\(_{0.35}\)) in stoichiometric amounts prior to spin-coating, we tuned the NCs to have a composition of CsPb(Cl\(_{0.65}/\text{Br}_{0.35}\)) (Figure S16, Supporting Information), which has a blue gain bandwidth of 473–480 nm. This wavelength range overlaps with the Γ point wavelength of $a_0 = 300 \text{ nm}$ Al

**Figure 3.** Angle-resolved transmission and lasing characteristics of quasi-propagating modes. a) Measured s-polarized band structure of the device along Γ–M with both $W_{\text{TE}}$-SLR and $W_{\text{TM}}$-SLR modes observed. b) Below-threshold photoluminescence measurements along Γ–M. c) Above threshold, lasing is observed over the energy range (27 meV) that the gain bandwidth overlaps with the $W_{\text{TM}}$-SLR bands.
NP square lattices at \((k_x, k_y) = (0, 0)\) as well as the quasi-propagating modes along \(\Gamma-X\) and \(M-X\) directions (Figure 4a). The cone intersections in reciprocal space converted into real-space polar coordinates show the expected patterns (Figure 4b). Lasing was observed at \(\approx 478\) nm (Figure S17, Supporting Information) from both the \(\Gamma\) point at \((\theta, \phi) = (0^\circ, 0^\circ)\) (Figure 4c, inset) and quasi-propagating modes with beams at \(\theta \approx 60^\circ\) and along the arms of the cross near \(\theta = 0^\circ\) (Figure 4c). Notably, lasing via biaxially confined modes and quasi-propagating modes can occur simultaneously. Additional experiments with perovskite NC compositions that emit at other wavelengths (e.g., \(\approx 500\) nm) also showed lasing emission patterns confirmed by the model (Figure S18, Supporting Information). By demonstrating lasing across different wavelength ranges and directions, we also distinguish quasi-propagating modes from accidental bound-states-in-the-continuum that support feedback only at specific wavevectors along the high-symmetry directions.\(^{[59,60]}\)

To extend quasi-propagating lasing to include coupling between the higher \((\pm 1, \pm 1)\) diffraction orders, we integrated CsPbBr\(_3\) NC with Al NP square lattices with a larger periodicity \(a_0 = 350\) nm. In this scenario, because the mode cones intersect past the \(\Gamma\) point and approach the \(X\) point, quasi-propagating modes have energies that are between \(\Gamma\) and \(X\).\(^{[61,62]}\) These quasi-propagating modes are comprised of four families of coupled diffraction orders: \((-1,0)/(0,-1)\) along \(\Gamma-M\); \((0,1)/(0,-1)\) and \((1,1)/(1,-1)\) along \(\Gamma-X\); and \((0,1)/(1,1)\) along \(X-M\) (Figure 4d). Sixteen lasing beams are distributed symmetrically about the rotational axis of the square lattice (Figure 4e,f and Figure S17, Supporting Information). The different beam profiles and orientations are from a combination of two factors: i) mode cones that couple at obtuse angles (e.g., \((0,1)/(0,-1)\)) intersect over a larger span of wavevectors for a given energy range compared to those at acute angles (e.g., \((0,1)/(1,1)\)); and ii) beams emitted at larger \(\theta\) have larger \(\Delta \theta\) because of the sinusoidal relationship between \((k_x, k_y)\) and \((\theta, \phi)\).\(^{[54]}\)

Finally, to demonstrate quasi-propagating modes in NP lattices with a different symmetry, we fabricated devices of Al NP hexagonal lattices and CsPbBr\(_3\) NCs. To match the reciprocal lattice constant of square lattices with \(a_0 = 300\) nm, we used a hexagonal lattice with periodicity \(a_0 = 346\) nm. Here, two sets of hexagonally distributed quasi-propagating modes are predicted, with those at smaller \(|k_x|\) along \(\Gamma-M\) and those at larger \(|k_x|\) along \(\Gamma-K\) (Figure 4g). Accordingly, optical feedback from these modes resulted in two concentric sets of lasing beams that...
3. Conclusion

We have demonstrated lasing from quasi-propagating modes along the high-symmetry directions of periodic plasmonic lattices. By optimizing both the gain material and lattice cavity, we showed that uniaxial feedback is sufficient to achieve lasing, resulting in multiple beams where each beam contained a range of wavelengths determined by the gain bandwidth. Quasi-propagating lasing characteristics are important in emerging technologies such as optical analog computing,[63] multichannel optical communication,[64] and portable 3D spatial sensing,[65,66] where multiplexed, chromatic lasers are desired. By generalizing wave behavior in periodic lattices, our work provides a strategy not only for designing integrated lasers in compact devices but also opens prospects for using high symmetry directions for band engineering in optical and phononic materials.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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