Room-Temperature Near-Infrared High-Q Perovskite Whispering-Gallery Planar Nanolasers

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ABSTRACT: Near-infrared (NIR) solid-state micro/nanolasers are important building blocks for true integration of optoelectronic circuitry.1 Although significant progress has been made in III–V nanowire lasers with achieving NIR lasing at room temperature,2–4 challenges remain including low quantum efficiencies and high Auger losses. Importantly, the obstacles toward integrating one-dimensional nanowires on the planar ubiquitous Si platform need to be effectively tackled. Here we demonstrate a new family of planar room-temperature NIR nanolasers based on organic–inorganic perovskite CH3NH3PbI3−aXa (X = I, Br, Cl) nanoplatelets. Their large exciton binding energies, long diffusion lengths, and naturally formed high-quality planar whispering-galley mode cavities ensure adequate gain and efficient optical feedback for low-threshold optically pumped in-plane lasing. We show that these remarkable wavelength tunable whispering-gallery nanolasers can be easily integrated onto conductive platforms (Si, Au, indium tin oxide, and so forth). Our findings open up a new class of wavelength tunable planar nanomaterials potentially suitable for on-chip integration.

KEYWORDS: Organic–inorganic perovskites, whispering gallery mode lasing, near-infrared lasers, nanoplatelets, wavelength tunable, on-chip integration

Semiconductor nanostructures (nanowires, nanoribbons, and so forth) are highly attractive gain media for applications as miniaturized solid-state laser components in integrated optoelectronic chips.1 However, as the laser size approaches subwavelength dimensions, the lasing gain threshold gth ∝ Γ in R−1 increases dramatically because of the low mode reflectivity R at the nanoscale end-facets and the high mode confinement factor Γ.5,6 In addition, the increased surface states further decrease the gain coefficient.7 In a variety of technologically important semiconductor nanostructures with high quantum efficiencies and large exciton binding energies (CdS, ZnO, GaN, and so forth), the gain is sufficient to compensate the losses for sustained visible and ultraviolet lasing at room temperature.8,9 Nevertheless, the development of solid-state room-temperature near-infrared nanolaser is still hampered by the low quantum efficiency of the gain materials (GaAs, CdTe, InP, and so forth) and small exciton binding energies (typically ∼5–6 meV, which are much smaller than the thermal kinetic energy of ∼26 meV at room temperature).2–4,10,11 Considerable efforts have been made to improve the quantum efficiency of the gain materials by surface passivation12 and to enhance the cavity quality via introducing whispering-gallery-mode (WGM) cavity, Bragg-reflector, and so forth.13 However, in GaAs nanowires the quantum efficiency is still below 1% because of the large surface areas and highly efficient nonradiative Auger recombination pathways.1,4,14 As a consequence, room-temperature near-infrared nanowire lasing is always realized through a compromise of the mode confinement, that is, the thickness of nanowire is up to ∼430 nm or larger,4,14 making it highly challenging for integration onto planar on-chip circuitry. It is therefore imperative to look for an entirely new class of more efficient solid-state near-infrared planar gain media compatible with the ubiquitous planar Si technologies.

Recently, a family of methylammonium lead halide perovskites CH3NH3PbI3−aXa (X = I, Br, Cl) (0 ≤ a ≤ 3) has attracted considerable attention for its breakthrough in improving solar-cell efficiency.15–18 These organic–inorganic semiconductor perovskites exhibit an energy gap around ∼770 nm at room temperature, large exciton binding energy (∼20 meV), long exciton diffusion length (∼100 nm), and lifetime (∼8 ns).18 A recent work in these perovskites thin films suggests that they also possess excellent optical gain properties.

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in a wide range of wavelengths. In this work, relatively high quantum yield for the perovskites (~17% for CH₃NH₃PbI₃) was proved as well as a more efficient stimulated emission over Auger process. Furthermore, through introducing distributed Bragg reflector (DBR) cavity, room-temperature lasing was realized in the CH₃NH₃PbI₃Cl perovskite that has a particularly long carrier lifetime and quantum yield (>30%) among the present perovskite serials. However, the laser device has bulk size in three dimensions. Attempts on achieving photonic and polariton lasing in these perovskites microcavities, particularly as nanoscale lasers, started several decades ago but with very limited progress. The reason may stem from the insufficient gain to compensate the loss due to (1) the lack of a high-quality cavity and (2) insufficient quantum efficiency limited by crystalline defects. These two disadvantages can be mitigated in crystalline perovskite nanoplatelets grown on mica substrates by chemical vapor methods shown in this work. Well-defined polygonal CH₃NH₃PbI₃-Xₐ (X = I, Br, Cl) perovskite WGM nanocavities are naturally formed during the growth process and crystalline quality is remarkably improved. The mode reflectivity at the end-facets can be large as ~85–95% because of the relief of confinement in the mode guided direction and the total internal reflectivity inside the WGM cavity. The multireflectance process further improves the optical feedback. In the family of the unique perovskite nanoplatelets, we have demonstrated strong optical-pumped room-temperature near-infrared lasing with low thresholds and wide mode-tunability. We further explore the possibility of direct integration of the nanoplatelet laser devices to other conductive substrates.

The CH₃NH₃PbI₃-Xₐ (X = I, Br, Cl) perovskites (Figure 1a) are converted from lead halide platelets grown on muscovite mica (see Methods). The as-grown lead halide shows well-defined triangular or hexagonal platelets with nanoscale thickness (10–300 nm) and edge length of several to tens of micrometers. The geometries are maintained after the conversion to perovskites as displayed in the optical images (Figure 1b–d, top panel). The perovskites show rich color dependent on the thickness, which can be correlated from the optical images and atomic force microscopy (AFM) data (Figure 1b–d, middle panel). The average surface roughness of perovskites is measured to be around 5 nm, which is perfectly flat in optical level. The absorption (Abs) and photoluminescence (PL) spectra are measured on individual nanoplatelets. The energy band gap of CH₃NH₃PbI₃, CH₃NH₃PbI₃Br, and CH₃NH₃PbI₃Cl are 765, 764, and 767 nm, respectively.

Figure 1. Chemical-vapor-deposited methylammonium lead halide perovskite nanoplatelets. (a) Schematics of atomic structures of threedimensional CH₃NH₃PbI₃-Xₐ (X = I, Br, Cl) perovskites. (b–d) Morphological and electronic band-edge characterizations of CH₃NH₃PbI₃ (b), CH₃NH₃PbI₃Br (c), and CH₃NH₃PbI₃Cl (d) nanoplatelets grown on mica substrates, respectively. Upper panel: optical images. The nanoplatelets show planar, well-defined, polygonal structures with an edge length of 5–50 μm and a thickness of 20–300 nm. The angles between the polygonal edges are 60 or 120°, consistent with the atomic structures of the perovskites. Middle panel: Cross-section view recorded by atomic force microscope. The nanoplatelet thickness corresponds to the optical contrast or distinguished color in optical image (inset). Bottom panel: absorption (olive) and photoluminescence (pink) spectra taken from individual nanoplatelets. The energy band gap of CH₃NH₃PbI₃, CH₃NH₃PbI₃Br, and CH₃NH₃PbI₃Cl are 765, 764, and 767 nm, respectively.
Figure 2. Lasing characterizations of perovskites whispering-gallery-mode nanocavities. (a) Schematic of optical setup. A pulsed laser (400 nm, 150 fs, 1 kHz) excites the sample from the top globally. The spot size is large enough to cover the whole cavity to ensure high pumping injection efficiency and reduce the local heating effect. The NIR photoluminescence from perovskites waveguides inside the cavities confined by their planar polygonal structures. (b) Far-field optical image of two typical CH3NH3PbI3 nanoplatelets under the illumination of white light (upper panel) and incidence laser (bottom panel). Clear diffraction patterns can be seen inside the whispering-gallery-mode cavity. (c) The evolution from spontaneous emission (SE, ~768 nm, peak center indicated by red arrow) to lasing (whispering-gallery modes indicated by blue arrows) in a typical CH3NH3PbI3 triangular nanoplatelet (optical image inset; thickness, 150 nm; edge length, 32 μm). The pumping fluence (Pfl) from low to high is 14 (gray), 40.6 (dark yellow), 42 (red), and 51 (olive) μJ/cm^2, respectively. Five near-infrared lasing modes are resolved at λ = 776.7, 779.2, 781.8, and 784.3 nm (Δλ = 2.6 nm, respectively). The full width at half-maximum (FWHM) of a lasing mode is λwhlm ~ 1.2 nm, which includes the instrument resolution broadening. The Q-factor is ~650, evaluated by Q = λ/λwhlm. Left inset: integrated output emission (Pout) over the whole spectra range as a function of pumping fluence (Pfl) using log—log scale. The experiment data (red points) is well fitted by distinguished functions (green line) with a knee at Pfl = 37 μJ/cm^2 (lasing threshold Pth). Right inset: Photoluminescence decay curve below (pink, SE) and above (dark green, lasing) the threshold. The SE average lifetime is 2.6 ns. Two lifetimes under lasing action are deduced out as ~80 ps (dominant) and 846 ps. (d) Parallel steady-states SE, lasing and time-resolved photoluminescence measurement on CH3NH3PbI3Br, with similar size. The pumping fluence Pfl is 87 (gray), 135 (dark yellow), 154 (red), and 175 (olive) μJ/cm^2, respectively. The lasing threshold Pth is ~128 μJ/cm^2. λwhlm ~ 0.9 nm for whispering-gallery mode λ = 785.6 nm and the corresponding Q-factor is ~900. The average SE and dominant lasing lifetime is 7.8 ns and 75 ps, respectively. Both spectra in the insets (c,d) are normalized using the highest intensity.

(For example, P = 14 μJ/cm^2, gray curve, Figure 2c), a broad spontaneous emission band centered at 768 nm with a full width at half-maximum (fwhm) of Δλwhlm ~ 54 nm can be seen (Supporting Information Figure S2). When the pump fluence increases to be 40.6 μJ/cm^2 (dark yellow curve, Figure 2c), several sharp peaks centered at λ = 776.7, 779.2, 781.9, 784.3, and 786.8 nm with a λwhlm of ~1.2 nm appear over the spontaneous emission band. The peak line width is comparable with that of reported in room-temperature nanowire and suspended WGM microdisk NIR lasers, suggesting the occurrence of lasing action. Each peak corresponds to one whispering-gallery cavity mode. The spacing between two adjacent peaks Δλ is almost the same, indicating the same waveguiding origin of the modes. As the pump fluence is further increased (P = 42 and 51 μJ/cm^2), the lasing intensity increases correspondingly and the lasing mode exhibits a slight blueshift. The detailed waveguide type and lasing mechanism will be discussed later. The log—log scale plot of emission output (Pout) versus pumping fluence shows a ~S-like curve, which can be well fitted with two linear functions in the spontaneous emission and lasing regimes (Figure 2c, left inset). Furthermore, a lasing threshold of Pth ~37 μJ/cm^2 is deduced, which is in good agreement with the appearance of the narrow peaks around the pumping fluence. Time-resolved PL study is also conducted (Figure 2c right panel). Below the threshold, the PL decay curve shows a long average lifetime of ~2.6 ns, indicative of the characteristics of the spontaneous emission, while an ultrafast decay channel with a lifetime of ~80 ps (limited by the system response of the streak camera over the time window) dominates above the threshold pumping due to the occurrence of exciton population inversion and lasing action (Supporting Information Figure S3).
All the steady-state and time-resolved PL/lasing studies are performed using the CH$_3$NH$_3$PbI$_3$-Br samples (Figure 2d) and CH$_3$NH$_3$PbI$_3$Cl, (Supporting Information S4). Room-temperature near-infrared lasing is also achieved with a threshold of $\sim$128 $\mu$J/cm$^2$. Several lasing peaks are observed around 785 nm with mode spacing consistent with each other, also suggesting the dominance of WGMs in the perovskites. Although the threshold of CH$_3$NH$_3$PbI$_3$-Br is higher than the CH$_3$NH$_3$PbI$_3$, for the similar size the fwhm of its lasing mode is smaller ($\sim$0.9 nm) thus the cavity quality factor ($Q \sim$900) is somewhat larger (Figure 2d). Moreover, our measurement in many CH$_3$NH$_3$PbI$_3$-Br nanoplatelets suggests that lasing with fewer modes is readily achieved, suggesting the potential of single-mode lasing, although the mechanism deserves further in-depth investigations.

Optical mode simulations are performed to further understand the lasing in the perovskites. Figure 3a shows the photoluminescence image of an individual hexagonal CH$_3$NH$_3$PbI$_3$ nanoplatelet above the lasing threshold. The lasing only couples out at the cavity edge, suggesting a good mode confinement in the plane of the nanoplatelets, yielding an in-plane emission. Figure 3b,c shows the absolute electric field distribution inside the hexagonal nanoplatelet (thickness, 150 nm; edge length, 10 $\mu$m) when the transverse magnetic (TM) and transverse electric (TE) modes dominates, respectively. In the two cases, the optical fields are well-confined inside the cavities and reflected between the polygonal facets to construct WGM waveguides. With decreasing nanoplatelet thickness, the effective index of the two photonic modes decreases, yielding a higher radiative loss or even the cutoff of the photonic modes, while TM mode has a large effective index thus a lower lasing threshold is anticipated. Experimentally, the lasing cutoff thickness at room temperature is $\sim$70 nm, while this value can be pushed to $\sim$40 nm at lower temperature ($\sim$77 K) because of the higher optical gain (Supporting Information S5).

The cavity resonant mode $\lambda$ and the corresponding mode number $N$ are further calculated, which agrees well with the theoretical value, confirming the occurrence of WGM lasing (Figure 3d, upper panel) (see Methods). When the pumping fluence is close to the lasing threshold (40.6 $\mu$J/cm$^2$), only TM-mode achieves lasing because of the lower gain threshold (Supporting Information Figure S5). As the pumping fluence increases from 40.6 to 50.4 $\mu$J/cm$^2$, the lasing of more resonant modes is realized because of the achievement of threshold gain at a high carrier-density. Additional lasing peaks appear near the TM modes (indicated by arrows, Figure 3d, bottom panel), which is identified as TE mode. Besides, the lasing modes (that is, 793.1 nm) are slightly blueshifted (that is, 792.6 nm), which may be due to a band-filling mode or refractive index variation (Figure 3d, bottom panel).11,25

For the nanoplatelets of different sizes, the lasing modes can be tuned due to the intrinsic self-absorption of the excitons. As the edge length $L$ increases from 28, 32, 38 to 47 $\mu$m (thickness of $\sim$150 nm ±10 nm), the highest optical gain area where lasing can be realized moves to the lower energy region (Figure 3e). The strongest lasing modes of the nanoplatelets are extracted and show an exponential decrease function of the edge length according to the self-absorption equations (Figure 3f).26 The limit of the lasing modes occurs at $\sim$790 nm as deduced from the fitting, which is close to the Urbach tail near

Figure 3. Whispering-gallery mode analysis of the perovskite nanoplatelet laser. (a–c) Far-field image (a), simulated field distributions at resonant cavity mode (b: transverse magnetic mode; c: transverse electric mode) of typical hexagonal CH$_3$NH$_3$PbI$_3$ nanoplatelets. The thickness and edge length is 150 nm and 32 $\mu$m. (d) Lasing spectra of hexagonal CH$_3$NH$_3$PbI$_3$ nanoplatelets (upper panel) and the lasing mode evaluation as pumping fluence (bottom panel). Hexagonal nanoplatelet: thickness $\sim$110 nm, edge length $\sim$33 $\mu$m. The optical image is shown in the inset and the lasing pumping fluence is indicated near each curve. The lasing mode and respective mode number are calculated and indicated in the spectra, which are in good agreement with theoretical result. The lasing modes blueshift as the pumping fluence increases. (e) Lasing spectra is dependent on the edge length of a triangular CH$_3$NH$_3$PbI$_3$ whispering-gallery cavity. The edge length is 47, 38, 32, and 28 $\mu$m (also indicated), respectively, from upper to bottom. The thickness is $\sim$150 nm. The lasing wavelength redshifts and mode spacing decreases with the increase of edge length. (f) The lasing mode and respective mode number are further calculated, which agrees well with the theoretical value, confirming the occurrence of WGM lasing (Supporting Information S5). As the pumping fluence is close to the lasing threshold (40.6 $\mu$J/cm$^2$), only TM-mode achieves lasing because of the lower gain threshold (Supporting Information Figure S5). As the pumping fluence increases from 40.6 to 50.4 $\mu$J/cm$^2$, the lasing of more resonant modes is realized because of the achievement of threshold gain at a high carrier-density. Additional lasing peaks appear near the TM modes (indicated by arrows, Figure 3d, bottom panel), which is identified as TE mode. Besides, the lasing modes (that is, 793.1 nm) are slightly blueshifted (that is, 792.6 nm), which may be due to a band-filling mode or refractive index variation (Figure 3d, bottom panel).11,25

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the bottom of the absorption edge (Figure 1b), thus further validating the self-absorption dependent lasing mode. Besides, the Q-factor (calculated by $\lambda/\lambda_{\text{FWHM}}$) increases linearly with the edge length (from 650 to 1320). For an m-faceted polygonal WGM cavity, the Q-factor of a resonant mode $\lambda$ is proportional to $L/\lambda$. $\lambda$ can be considered as a constant; therefore the Q-factor has a linear relationship with the edge length (Figure 3f). Although the perovskite nanolaser platelet has a subwavelength thickness (40–150 nm), the Q-factor of the on-substrate laser is relatively higher than reported in suspended microdisk WGM, nanowire, and the recent polycrystalline perovskite thin film DBR NIR lasers, suggesting that the large cavity scattering and radiation loss is well overcompensated by the high optical gain of the materials.

To prove the potential of integration onto optoelectronic chips, we demonstrate the functional lasing onto other conductive bases such as Si, Au, and indium tin oxide (ITO). Because our perovskites are grown on a transparent insulating substrate, the integration can be accomplished via a simple reversing of the substrate onto the bases without any releasing process or change of pumping method (Figure 4a,b) with the same CH$_3$NH$_3$PbI$_3$ hexagonal nanoplatelets (thickness, 110 nm; edge length, 33 μm). Room-temperature lasing is realized on all four substrates (Figure 4c). Because the thickness of CH$_3$NH$_3$PbI$_3$ is smaller than its diffraction length (∼200 nm), the spontaneous emission lifetimes decrease gradually from mica (2.89 ns), Si (2.6 ns), ITO (1.9 ns), and Au (1.2 ns) to the spontaneous emission lifetimes decrease gradually from mica (2.89 ns), Si (2.6 ns), ITO (1.9 ns), and Au (1.2 ns). The threshold of the four types of nanolaser is almost the same as $\sim$40 μJ/cm$^2$. The wavelength and relative intensity of the lasing modes are almost the same. (d) SE decay function with average time constants of mica (2.89 ns), Si (2.6 ns), ITO (1.9 ns), and Au (1.2 ns).

In summary, we have realized room-temperature near-infrared solid-state nanolaser devices using the organic–inorganic lead halide perovskite nanoplatelets. The large exciton binding energy, long diffusion length, as well as promising quantum yields of the family leads to strong, stable, and well-controlled lasing actions. The whispering-gallery type planar perovskite nanolaser devices have pronounced optical gain and tunable optical modes, which can be potentially integrated with the existing Si technologies. Our research opens alternative routes beyond III–V nanostructures in achieving near-infrared solid state nanolasing and will inspire more designs of low-threshold near-infrared nanolasers pumped optically and electrically.

**METHODS**

**Perovskite Synthesis.** Either PbI$_2$, PbBr$_2$, or PbCl$_2$ powder (99.99%, Aldrich) was used as a single source and placed into a quartz tube mounted on a single zone furnace (Lindberg/Blue M TFS503S-1). The fresh-cleaved muscovite mica substrate (1 cm × 3 cm) was precleaned by acetone and placed in the downstream region inside the quartz tube. The quartz tube was first evacuated to a base pressure of 2 mTorr, followed by a 30 sccm flow of high purity Ar premixed with 5% H$_2$ gas. The temperature and pressure inside the quartz tube were set and stabilized to desired values for each halide (380 °C and 200 Torr for PbI$_2$; 350 °C and 75 Torr for PbBr$_2$; and 510 °C and 200 Torr for PbCl$_2$). In all cases, the synthesis was done within 20 min and the furnace was allowed to cool down to ambient temperature naturally. After that, pregrown lead halide platelets were thermally intercalated with methylammonium iodide in a fresh quartz tube (using the same furnace system). The platelets were placed in the downstream region while the methylammonium iodide was placed in the center of the tube. The intercalation was carried out at 115 °C under a pressure of...
40 Torr with a 30 sccm flow of high purity Ar or N₂ gas for 1 h to fully convert lead halides to perovskites.

**Steady-State Photoluminescence and Absorption Spectroscopy.** Steady-state PL spectra were conducted at room temperature using a micro-Raman spectrometer (Horiba-JY T64000) in a backscattering configuration. A series of laser lines 633 nm (solid state laser) were used as the excitation sources. The backscattered signal was collected through a 100X objective and recorded by a liquid nitrogen cooled charge coupled device detector. The absorption spectra of individual perovskite nanoplatelets were conducted at room temperature using a commercial microtransmission/absorption spectrometer (Craic 20/20). The light from a Xe lamp is focused onto the sample normally from bottom. The transmitted light is collected by a reflective objective (36X , numerical aperture: 0.4), and spectrally analyzed by a monochromator. Apertures of suitable size are used to acquire transmission light from targeted area.

**Lasing Spectroscopy/Image and Time-resolved Photoluminescence Spectroscopy.** The 400 nm excitation pulses were generated by frequency doubling the 800 nm output (with a BBO crystal) from the Coherent Libra regenerative amplifier (50 fs, 1 kHz, 800 nm) that is seeded by a Coherent Vitesse oscillator (50 fs, 80 MHz). The pumping source are introduced into a microscope (Nikon LV100) and focused onto samples via a 20X objective (Nikon, numerical aperture: 0.4). To ensure sufficient energy injection, the laser spot diameter is focused to ~45 μm. The backscattered emission is collected by the same objective. For collecting the laser image, the emission signals are imaged on a CCD camera with a long-pass filter to block the laser line. For lasing spectroscopy, the emission signal from area of ~5 μm × 5 μm goes through an aperture and analyzed by a spectrometer (Princeton Instrument SP2300i) equipped with a TE-cooled charge coupled device detector. For time-resolved photoluminescence spectra, the PL emission was collected in a standard backscattering geometry and dispersed by a 0.25 m DK240 spectrometer with 150 g/mm grating. The emission was time-resolved using an Optoscope Streak Camera system that has an ultimate temporal resolution of ~50 ps.

**Numerical Calculations.** The photonic eigenmodes in perovskites and corresponding effective index are calculated by finite element method (FEM, COMSOL). Only the fundamental transverse magnetic and transverse electric have effective indices larger than mica substrate. The wavelength-dependence refractive index of mica and perovskites are taken from previous literatures (details see Supporting Information Figure S5). The field distribution of the whispering-gallery waveguide modes are further analyzed by a two-dimensional mode solution method using the effective indices. The combined one-dimensional and two-dimensional methods are previously used in Nat. Mater. 2011, 10, 110–113.

The cavity resonant mode λ and corresponding mode number N are deduced by the equation \( N = \frac{3(3n_D D^{1/2})}{2\lambda} \tan^{-1} \left[ \frac{n_J (3n_J^2 - 4)^{1/2}}{\lambda} \right] \) (Phys. Rev. A 2005, 72, 023806), where D is the diameter of the circle that circumscribes the polygon cavities, and \( n_J \) is refractive index at the resonant wavelength.

**ASSOCIATED CONTENT**

**Supporting Information**

Description of experimental results on exciton binding energy, room-temperature lasing spectra, time-resolved emission spectra, and quantum efficiency estimation, the whispering gallery mode simulation. This material is available free of charge via the Internet at http://pubs.acs.org.

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**Author Contributions**

Q.Z., S.T.H., and Q.X. conceived the idea and designed the experiment. S.T.H. prepared the samples and performed optical absorption, photoluminescence spectroscopy measurements, and AFM measurements. Q.Z., X.F.L. and T.S. conducted the lasing and time-resolved photoluminescence measurement. Q.Z. and X.F.L. performed the simulation. All the authors cowrote the manuscript.

Q.Z., S.T.H., and X.L. contribute the same to the work.

**Notes**

The authors declare no competing financial interest.

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