

Enhancement of laser power-efficiency by control of spatial hole burning interactions

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1. THE STEADY-STATE AB-INITIO LASER THEORY FOR SPATIALLY INHOMOGENEOUSLY PUMPED LASERS

Here we provide a complete derivation of the results in the main text, along with additional details for basic properties of lasers that are pumped spatially inhomogeneously.

1.1. Gain-medium Independent Form of the Semiclassical Laser Equations

The semiclassical laser theory [1, 2] is sufficient to capture characteristics such as thresholds, frequencies and output intensities of a wide range of laser systems. This theory treats the electromagnetic fields classically and the lasing medium quantum-mechanically, leading to a coupled set of non-linear partial differential equations referred to as the Maxwell-Bloch (MB) equations,

$$\nabla^2 E^+ - \frac{1}{c^2} \ddot{E}^+ = \mu_0 \ddot{P}^+, \quad (\text{S1})$$

$$\dot{P}^+ = -(i\omega_a + \gamma_\perp)P^+ + \frac{g^2}{i\hbar} E^+ D, \quad (\text{S2})$$

$$\dot{D} = \gamma_\parallel [D_0(\vec{r}) - D] - \frac{2}{i\hbar} [E^+(P^+)^* - (E^+)^* P^+]. \quad (\text{S3})$$

Only the positively rotating field components, identified by a “+” in the superscript, appear in the equations above due to the Rotating Wave Approximation (RWA). Here c and μ_0 are the speed of light and permeability in vacuum, ω_a is the atomic transition frequency, $\gamma_\parallel$ and $\gamma_\perp$ are the inversion and polarization relaxation rates, $\hbar$ is the reduced Planck constant, and g is the transition dipole matrix element between the lasing levels.

The steady-state solution of these equations requires $\dot{D} \approx 0$ (i.e., the stationary inversion approximation), under which the following solution multi-periodic in time can be assumed:

$$E^+(\vec{r}, t) = \sum_{\mu=1}^N \Psi_\mu(\vec{r}) e^{-i\Omega_\mu t}, \quad P^+(\vec{r}, t) = \sum_{\mu=1}^N p_\mu(\vec{r}) e^{-i\Omega_\mu t}, \quad (\text{S4})$$

where N is the finite number of lasing modes at a given pump power. The polarization field amplitude can be obtained by inserting the ansatz for each field into Equation (S2) and using the orthogonality of the Fourier basis in time,

$$p_\mu(\vec{r}) = -i \frac{g^2}{\hbar} \frac{D(\vec{r})}{i(\omega_a - \Omega_\mu) + \gamma_\perp} \Psi_\mu(\vec{r}). \quad (\text{S5})$$

We then obtain an expression for the steady-state inversion,

$$D(\vec{r}) = \frac{D_0(\vec{r})}{1 + \sum_{\mu=1}^N \Gamma_{\mu} |\Psi_{\mu}(\vec{r})|^2}. \quad (\text{S6})$$

Plugging Equation (S6) into Equation (S5) and using Equation (S4) in (S1), we obtain the set of non-linear Helmholtz equations for the steady state amplitudes displayed in Eq. (1) in the main text:

$$[\nabla^2 + (\epsilon_c(\vec{r}) + \epsilon_g(\vec{r}))\Omega_{\mu}^2] \Psi_{\mu}(\vec{r}) = 0, \quad \vec{r} \in \text{cavity} \quad (\text{S7})$$

$$\epsilon_g(\vec{r}) = \frac{\gamma_{\perp}}{\Omega_{\mu} - \omega_a + i\gamma_{\perp}} \frac{D_0(\vec{r})}{1 + \sum_{\nu=1}^N \Gamma_{\nu} |\Psi_{\nu}(\vec{r})|^2}. \quad (\text{S8})$$

This is the fundamental equation underlying the Steady-state Ab-initio Laser Theory (SALT) [3–6]. It finds solutions for $\Psi_{\mu}(\vec{r})$ and the corresponding lasing frequencies Ω_{μ} in arbitrarily complex and open photonic environments. The parameter $D_0(\vec{r})$ represents a generalized pump, parametrizing the strength of the applied pump at position $\vec{r}$ in terms of the unsaturated inversion density generated at that point. This parametrization allows us to describe the laser dynamics independent of the particular gain mechanism, as long as two laser levels can be identified (see Sec. 2). The inversion, subject to being spatially modulated by, for example, a patterned contact, can be described by

$$D_0(\vec{r}) = D_0 f(\vec{r}), \quad (\text{S9})$$

where $f(\vec{r})$ is normalized as $\int_{\text{cavity}} f(\vec{r}) d\vec{r} = S$ and consequently D_0 measures the total power injected in terms of the average inversion density generated (averaged over the cavity area S) in the absence of gain saturation. For a selectively pumped scenario, such as the one discussed in the manuscript, we distinguish two cases here: For pumping methods which would focus the pump into the selected areas (i.e. an axicon lens as in Ref. [7] for optical pumping, or shaped contacts as in Ref. [8] for electrical pumping) S is the total area of the cavity. Else, as in the case of an SLM, S is the area of the pumped region). Unless stated otherwise, we will consider the former case. While the Maxwell-Bloch Equations (S1)-(S3) are traditionally used to simulate gain in a medium represented by incoherently pumped 2-level systems, our treatment of the pump is free of this restriction. For a specified gain medium, $D_0(\vec{r})$ (as well as $\gamma_{\perp}$ and $\gamma_{\parallel}$) can be reparametrized to model exact device characteristics (see Sec. 2).

To fully analyze open systems using Equation (S7), we expand each field component, oscillating at individual frequencies Ω_μ , into a set of biorthogonal and complete basis functions called the Constant Flux (CF) states [3],

$$\Psi_\mu(\vec{r}) = \sum_m a_m^\mu \phi_m(\vec{r}, \Omega_\mu). \quad (\text{S10})$$

The CF states are defined through the eigenvalue problem of the Laplace operator $\mathcal{L} = -\frac{1}{\epsilon_c(\vec{r})} \nabla^2$,

$$\mathcal{L} \phi_m(\vec{r}, \Omega_\mu) = \omega_m^2(\Omega_\mu) \phi_m(\vec{r}, \Omega_\mu), \quad (\text{S11})$$

and the frequency-dependent outgoing boundary conditions $\partial_r \phi_m(r, \Omega_\mu) = i\Omega_\mu \phi_m(r, \Omega_\mu)$ at $r = |\vec{r}| \rightarrow \infty$. $\omega_m \equiv \nu_m - i\kappa_m$ are the complex CF frequencies of this eigenvalue problem. The CF states satisfy the following biorthogonal relation,

$$\int_{\text{cavity}} d\vec{r} \epsilon_c(\vec{r}) \phi_m(\vec{r}, \Omega_\mu) \phi_n(\vec{r}, \Omega_\mu) = \delta_{mn}, \quad (\text{S12})$$

which differs from the power orthogonality relation of eigenstates of Hermitian operators. Below we discuss the case where $\epsilon_c(\vec{r}) = \text{const}$, and we rewrite the biorthogonal relation as

$$\int_{\text{cavity}} d\vec{r} \phi_m(\vec{r}, \Omega_\mu) \phi_n(\vec{r}, \Omega_\mu) = \delta_{mn} \quad (\text{S13})$$

for convenience.

Inserting the expansion (S10) in Equation (S7) and applying the biorthogonality (S13) results in a set of non-linear equations for the expansion coefficients $\{a_m^\mu\}$,

$$a_m^\mu = \frac{\Omega_\mu^2}{\omega_m^2 - \Omega_\mu^2} \frac{1}{\epsilon_c} \sum_q \int_{\text{cavity}} d\vec{r} \epsilon_g(\vec{r}) \phi_m(\vec{r}; \Omega_\mu) \phi_q(\vec{r}; \Omega_\mu) a_q^\mu. \quad (\text{S14})$$

This set of equations can be solved numerically for $\{a_q^\mu\}$ and the associated frequency Ω_μ for each lasing mode, accounting fully for interactions between all lasing modes. The results shown in Fig. 2 in the main text are obtained by solving this non-linear set of coupled equations for lasing amplitudes using the algorithm developed in Refs. [3, 6].

Below we also derive the semi-analytical result given in Ref. [5] that generalizes the well-known Haken-Sauermann [5] multi-mode laser equations to an open cavity, without expanding the spatial-hole burning interaction to the first order of the laser intensity (often referred to as the “third-order” approximation in terms of the amplitude of the lasing modes).

It requires an alternative treatment of Equation (S7): We move the term $\epsilon_g \Omega^2 \Psi_\mu$ to the right hand side and multiply both sides by $-(1 + \sum_{\nu=1}^N \Gamma_\nu |\Psi_\nu(\vec{r})|^2)/\Omega_\mu^2$, which results in

$$-\left(1 + \sum_{\nu=1}^N \Gamma_\nu |\Psi_\nu(\vec{r})|^2\right) \left[\frac{\nabla^2}{\Omega_\mu^2} + \epsilon_c\right] \Psi = \frac{D_0 \gamma_\perp}{\Omega_\mu - \omega_a + i\gamma_\perp} \Psi_\mu. \quad (\text{S15})$$

We then invoke the expansion (S10), operate on both sides of this equation with $\int_{\text{cavity}} d\vec{r} \phi_m(\vec{r}; \Omega_\mu)$ and use the biorthogonal relation (S13). The result is

$$\sum_q \eta_{q\mu} \left[\delta_{mq} + \sum_{\nu ss'} \Gamma_\nu \chi_{(mq)(ss')}^{\mu\nu} a_s^\nu a_{s'}^{\nu*} \right] a_q^\mu = \frac{D_0 \gamma_\perp}{\Omega_\mu - \omega_a + i\gamma_\perp} a_m^\mu, \quad (\text{S16})$$

where $\chi_{(mq)(ss')}^{\mu\nu} = \int_{\text{cavity}} d\vec{r} (\phi_{m\mu} \phi_{q\mu})(\phi_{s\nu} \phi_{s'\nu}^*)$, $\eta_{m\mu} = \frac{\omega_m^2 - \Omega_\mu^2}{\Omega_\mu^2} \epsilon_c$, and $\phi_{m\mu} \equiv \phi_m(\vec{r}; \Omega_\mu)$.

Equation (S16) is equivalent to Equation (S14) and accounts for the full modal interactions without the third-order approximation. Its advantage is the straightforward derivation of a set of *linear* equations in terms of the intensity of the lasing modes. To this end, we apply the single-pole approximation (SPA) [5] for each lasing mode $\Psi_\mu(\vec{r})$, e.g. $\Psi_\mu(\vec{r}) \approx a_\mu \phi_{m\mu}(\vec{r})$ and $\Psi_\nu(\vec{r}) \approx a_\nu \phi_{n\nu}(\vec{r})$, which is a good approximation for high- Q modes. With the SPA, the subscript m_μ, n_ν can be replaced by μ, ν without ambiguity, and Equation (S16) reduces to the following form:

$$\sum_{\nu=1}^N \text{Re}[\chi_{\mu\nu}] \Gamma_\nu I_\nu \approx \frac{D_0}{D_{th}^\mu} - 1, \quad (\text{S17})$$

where $\chi_{\mu\nu}$ is short for $\chi_{(m_\mu m_\mu)(n_\nu n_\nu)}^{\mu\nu}$, $I_\mu \equiv |a_\mu|^2$, and $D_{th}^\mu = \text{Re}[\lambda_\mu]^{-1} \equiv \text{Re}[\gamma_\perp / \eta_\mu (\Omega_\mu - \omega_a + i\gamma_\perp)]^{-1}$ is the threshold pump power for mode μ . We note that for high- Q modes $\chi_{\mu\nu}$ and λ_μ are approximately real, and hence the “Re[.]” notation is left out in Ref. [5]. As noted, Equation (S17) is the generalization of the well-known Haken-Sauermann [5] multi-mode laser equations to an open cavity, without the third-order approximation.

In the single-mode regime, Equation (S17) is further reduced to

$$I_\mu \approx \frac{1}{\Gamma_\mu \text{Re}[\chi_{\mu\mu}]} \left(\frac{D_0}{D_{th}^\mu} - 1 \right). \quad (\text{S18})$$

Equation (4) in the main text is derived by taking into account $|\Omega_\mu - \omega_a| \ll \gamma_\perp$ for a lasing mode close to the gain center ω_a , and the expression for D_{th}^μ leads to

$$\frac{\epsilon_c \omega_\mu^2}{\Omega_\mu^2} \approx \epsilon_c - i D_{th}^\mu. \quad (\text{S19})$$

For convenience, we denote the CF frequency $\omega_\mu \equiv \nu_\mu - i\kappa_\mu$, similar to the corresponding complex resonance used in the main text to define the outcoupling Q -factor in the absence

of absorption (i.e. $e_c = n_0^2 > 0$). It can be shown that for a high- Q mode these two corresponding frequencies are almost identical [3]. Equation (S19) leads to

$$\Omega_\mu \approx \sqrt{\nu_\mu^2 - \kappa_\mu^2} \approx \nu_\mu, \quad (\text{S20})$$

$$D_{th}^\mu \approx \frac{2n_0^2 \nu_\mu \kappa_\mu}{\Omega_\mu^2} \approx \frac{2n_0^2 \kappa_\mu}{\nu_\mu} = \frac{n_0^2}{Q_\mu} \equiv D_{out}^\mu \quad (\text{S21})$$

We note that the left hand side of Equation (S19) does not depend on whether the absorption is present; this can be seen from the definition of the CF states given by Equation (S11), where the product $\epsilon_c \omega_\mu^2$ is fixed for a given Ω_μ . Thus the right hand side of Equation (S19) should equal for $\epsilon_c = n_0^2$ and $(n_0 + in_1)^2$ as well, i.e.

$$n_0^2 - iD_{out}^\mu \approx (n_0 + in_1)^2 - iD_{th}^\mu, \quad (\text{S22})$$

the imaginary part of which gives $D_{th}^\mu \approx D_{out}^\mu + 2n_0 n_1$, i.e. Equation (4) in the main text.

If we do not make the approximation that $|\Omega_\mu - \omega_a| \ll \gamma_\perp$, a more accurate expression for the lasing frequency Ω_μ and threshold D_{th}^μ can be obtained at $D_0 = D_{th}^\mu$ where $I_\mu = 0$, i.e. from

$$\frac{D_0}{\epsilon_c} \frac{\gamma_\perp}{\Omega_\mu - \omega_a + i\gamma_\perp} \frac{\Omega_\mu^2}{\Omega_\mu^2 - \omega_\mu^2} + 1 = 0. \quad (\text{S23})$$

When absorption is absent, the result gives the familiar line-pulling formula

$$\Omega_\mu = \frac{\kappa_\mu}{\kappa_\mu + \gamma_\perp} \omega_a + \frac{\gamma_\perp}{\kappa_\mu + \gamma_\perp} \nu_\mu, \quad (\text{S24})$$

which becomes the leading order approximation $\Omega_\mu \approx \nu_\mu$ given previously when $\kappa_\mu \ll \gamma_\perp$, and the threshold

$$D_{th}^\mu = \frac{2n_0^2 \kappa_\mu}{\Omega_\mu} \left[1 + \frac{(\omega_a - \nu_\mu)^2}{(\kappa_\mu + \gamma_\perp)^2} \right] \approx D_{out}^\mu \left[1 - \frac{1}{2Q_\mu} \frac{\omega_a - \nu_\mu}{\kappa_\mu + \gamma_\perp} + \frac{(\omega_a - \nu_\mu)^2}{(\kappa_\mu + \gamma_\perp)^2} \right], \quad (\text{S25})$$

which becomes D_{out}^μ defined previously when $Q_\mu \gg 1$ and $|\omega_a - \nu_\mu| \ll (\kappa_\mu + \gamma_\perp)$.

In the next section we will derive the optimal Q -factor given by Equation (5) in the main text using Equation (S18) above and the outcoupling efficiency C_μ to be computed.

1.2. Mode out-coupling and flux

In this section, we first compute the “out-coupling coefficient” for a circular cavity using a general procedure that can be extended to other geometries. In the single-pole approximation,

the threshold CF states of a circular cavity for each mode μ are defined as:

$$\phi_\mu(r, \phi) = \begin{cases} \frac{1}{\zeta_\mu} J_m(n_c \omega_\mu r) e^{im\phi}, & r < R, \\ \frac{1}{\zeta_\mu} \frac{J_m(n_c \omega_\mu R)}{H_m^+(\Omega_\mu R)} H_m^+(\Omega_\mu r) e^{im\phi}, & r > R. \end{cases} \quad (\text{S26})$$

where $n_c = \sqrt{\epsilon_c}$ and $\zeta_\mu = [2\pi \int_{\text{cavity}} J_m^2(n_c \omega_\mu r) r dr]^{1/2}$ is a normalization constant that follows from the biorthogonality relation (S13). The outgoing power is expected to be proportional to the field energy stored inside the cavity. The associated proportionality constant emerges directly from the calculation of the total time-averaged flux (i.e., output power) from the cavity as measured at any $r > R$,

$$P_{\text{out}}(r) = \int_0^{2\pi} \int_{-Z/2}^{Z/2} \langle \vec{S}(r) \rangle \cdot \hat{r} \, d\theta \, dz \, \hat{r}, \quad (\text{S27})$$

where $\langle \vec{S}(r) \rangle = \frac{1}{2\mu_0} \text{Re}[\vec{E} \times \vec{B}^*]$ and Z is vertical spread of the beam. In the far-field, we approximate the outgoing Hankel function as follows,

$$H_m^+(\Omega_\mu r) = \sqrt{\frac{2}{\pi \Omega_\mu r}} e^{i(\Omega_\mu r + \gamma_m)}. \quad (\text{S28})$$

Here, $\gamma_m = -\frac{\pi m}{2} - \frac{\pi}{4}$. The electric field is given by

$$E(t) = E_0 \sum_{\mu} \Psi_\mu(r) e^{-i\Omega_\mu t}, \quad (\text{S29})$$

where $E_0 = \hbar \sqrt{\gamma_{\parallel} \gamma_{\perp}} / 2g$ is the natural units of the electrical field introduced in the main text [3]. Using the single-pole approximation and Equation (S28) yields the following expression for the electric field for the TM-polarized solutions,

$$\vec{E}_\mu(r, t) = \frac{E_0}{\zeta_\mu} \frac{J_m(n_c \omega_\mu R)}{H_m^+(\Omega_\mu R)} \sqrt{\frac{2}{\pi \Omega_\mu r}} e^{i(\Omega_\mu r + \gamma_m + m\phi)} e^{-i\Omega_\mu t} a_\mu \hat{z}, \quad (\text{S30})$$

$$= E_z(r, t) \hat{z}. \quad (\text{S31})$$

The following expression for the magnetic flux density $\vec{B}_\mu$ can be calculated using Maxwell's equations for $r \rightarrow \infty$ (Note: although we take $c = 1$ everywhere above, we include it here to avoid confusion regarding units),

$$\vec{B}_\mu(r, t) \approx -\frac{1}{c} E_z(r, t) \hat{\phi} \quad (\text{S32})$$

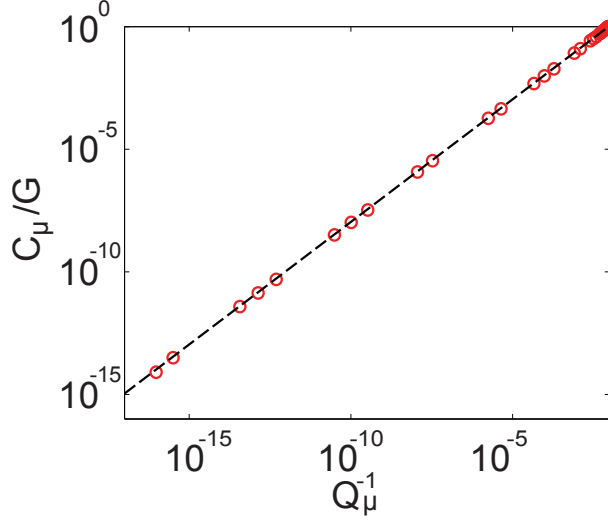


FIG. S1. Outcoupling efficiency C_μ as a function of the inverse of the outcoupling Q -factor. Modes with $m \leq 40$ in Figure 3(b) in the main text are used for this calculation. A linear dashed line $1.05 \times 10^4 Q_\mu^{-1}$ is shown as a comparison.

Here we neglect a non-zero component of $\vec{B}_\mu$ in the $\hat{r}$ direction because it is orthogonal to the flux in the radial direction. In addition, a subleading term that scales as $r^{-3/2}$ has been dropped in the expression above. Equation (S27) can now be evaluated exactly to yield,

$$P_{\text{out}}^\mu(r = \infty) = \frac{2Z}{\mu_0 \Omega_\mu} \left| \frac{E_0}{\zeta_\mu} \frac{J_m(n_c \omega_\mu R)}{H_m^+(\Omega_\mu R)} a_\mu \right|^2, \quad (\text{S33})$$

assuming that the variation of the fields in the vertical direction is negligible. This is the exact expression for the total power emitted from a mode μ of a circular disk cavity. Through SALT we get an exact expression for $|a_\mu|^2$, which is proportional to the energy stored in the cavity under steady-state conditions. The mode-dependent out-coupling coefficient for a disk cavity, as defined in the main text, is then

$$C_\mu = G \frac{\omega_a}{\Omega_\mu} \left| \frac{1}{\zeta_\mu} \frac{J_m(n_c \omega_\mu R)}{H_m^+(\Omega_\mu R)} \right|^2, \quad (\text{S34})$$

where $G = \frac{2ZE_0^2}{\mu_0 \omega_a}$.

Since C_μ is a measure of the power transfer rate from the intra-cavity intensity $|a_\mu|^2$ to the farfield, it should be proportional to the cavity decay rate κ_μ , or equivalently, the inverse of the outcoupling Q -factor Q_μ as shown in Fig. S1. Therefore, the output power P_{out}^μ is

proportional to the following quantity

$$\tilde{P}_{\text{out}}^\mu \equiv \frac{|a_\mu^2|}{Q_\mu} = \frac{1}{\Gamma_\mu \chi_{\mu\mu}} \left(\frac{\beta}{Q_\mu + Q_{\text{abs}}} - \frac{1}{Q_{\text{abs}}} \right), \quad (\text{S35})$$

where $\beta \equiv D_0/D_{\text{abs}}$ and $D_{\text{abs}} \equiv 2n_0n_1$ is defined in the main text. In the last step we have used Equation (S18) and Equation (4) in the main text. For a given cavity (and thus Q_{abs}) and pump power D_0 , the maximum output power is achieved when $\partial \tilde{P}_{\text{out}}^\mu / \partial Q_\mu = 0$, assuming that $\chi_{\mu\mu}$ and Γ_μ do not vary much among different modes we consider. This leads to the optical Q -factor given by Equation (5) in the main text. At this Q value, the maximum output power is given by

$$\tilde{P}_{\text{out}}^{\text{max}} = \frac{(\sqrt{D_0} - \sqrt{D_{\text{abs}}})^2}{n_0^2 \Gamma_\mu \chi_{\mu\mu}}. \quad (\text{S36})$$

1.3. Threshold Reduction Under Selective Pumping

If the pump has a nonuniform spatial profile $f(r)$, we obtain from Equation (S14)

$$D_{\text{out}}^\mu + D_{\text{abs}} \approx D_{\text{th}}^\mu \sum_q \frac{a_q^\mu}{a_m^\mu} \int_{\text{cavity}} d\vec{r} f(\vec{r}) \phi_m(\vec{r}; \Omega_\mu) \phi_q(\vec{r}; \Omega_\mu) \quad (\text{S37})$$

for $\Psi_\mu \rightarrow 0$ and to the leading order of n_1 . Invoking the single-pole approximation, i.e. keeping only the term proportional to

$$f_\mu \equiv \int_{\text{cavity}} f(\vec{r}) \phi_m^2(\vec{r}; \Omega_\mu) d\vec{r}$$

on the right hand side of Equation (S37), we derive

$$D_{\text{out}}^\mu + D_{\text{abs}} \approx D_{\text{th}}^\mu f_\mu, \quad (\text{S38})$$

from which the pump-focusing effect due to f_μ is readily seen. We note that f_μ is complex valued in general, but it is close to being real for high- Q modes.

2. SELECTIVE PUMPING: CASE STUDIES

In this section we discuss the implications of the results of the main text to (i) different types of gain media and (ii) resonator types. In Secs. 2.1 and 2.2, we discuss the gain-medium specific generalizations of the formulation of the laser theory described in Sec. 1. In Sec. 2.3, we discuss the power enhancement in a deformed resonator, and in Sec. 2.4, the case of a ridge cavity.

2.1. SALT for Selectively Pumped Quantum Cascade Lasers

The Maxwell-Bloch equations describe the pump $D_0(\vec{r})$ as the inversion density maintained at $\vec{r}$ due to an arbitrary pumping mechanism. As outlined in the general procedure above, this pump can be parametrized in terms of operational device parameters such as applied current and voltage. Here, we show this reparametrization for a Quantum Cascade (QC) laser accounting for the quantum transport of the charge carriers through the device.

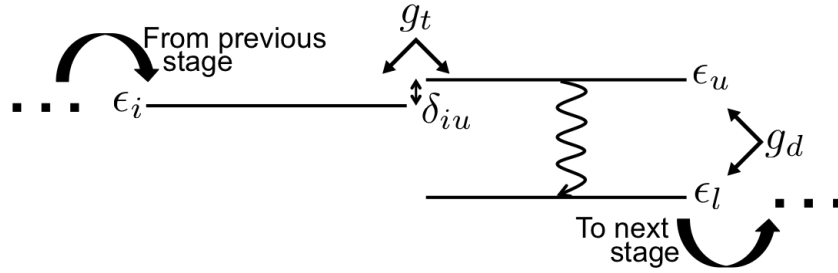


FIG. S2. A single stage of the cascade of 3-level systems used to model the QCL gain medium is shown. The angled arrows indicate coupling between levels, while curved arrows indicate transport of electrons from one stage to the next. The squiggly arrow between energy levels u and i indicates a radiative transition.

A QC gain medium can be represented as a cascade of three-level systems, each one consisting of an injector level (i), and upper (u) and lower (l) laser transition levels corresponding to energies ϵ_i , ϵ_u , and ϵ_l , respectively. The l level of the first three-level unit decays into the i level of the next three-level unit from where it tunnels into the laser transition quantum well and the process repeats (Fig. S2). The u level is detuned from the i level by a voltage-dependent detuning $\delta_{iu} = (\epsilon_i - \epsilon_u)/\hbar$. We employ a density-matrix approach to find the steady-state behavior of such a system as modeled by the following Hamiltonian,

$$H = \epsilon_i |i\rangle \langle i| + \epsilon_u |u\rangle \langle u| + \epsilon_l |l\rangle \langle l| + g_d E(t) (|u\rangle \langle l| + h.c.) + g_t (|i\rangle \langle u| + h.c.), \quad (\text{S39})$$

where $E(t)$ is the electric field, and g_d and g_t are the dipole moment and tunneling energy. The relaxation and pure dephasing dissipators, $\mathcal{D}_r$ and $\mathcal{D}_d$, are included in the equation of motion for the three-level density-matrix,

$$\dot{\rho} = -\frac{i}{\hbar} [H, \rho] + \mathcal{D}_r[\rho] + \mathcal{D}_d[\rho]. \quad (\text{S40})$$

The dissipators are described through the Lindblad superoperator,

$$\mathcal{D}_r[\rho] = \sum_{mn} c_{mn} \rho c_{mn}^\dagger - \frac{1}{2} \sum_{mn} (c_{mn}^\dagger c_{mn} \rho + \rho c_{mn}^\dagger c_{mn}), \quad mn = iu, ul \quad (\text{S41})$$

$$\mathcal{D}_d[\rho] = \sum_{mm} d_{mm} \rho d_{mm}^\dagger - \frac{1}{2} \sum_{mm} (d_{mm} \rho + \rho d_{mm}), \quad mm = ii, uu \quad (\text{S42})$$

and the collapse operators are defined as $c_{mn} = \sqrt{\Gamma_{mn}} \sigma_{mn}$ and $d_{mm} = \sqrt{\Gamma_m} \sigma_{mm}$, where $\sigma_{ij} = |j\rangle \langle i|$. Inter-band relaxation processes are indicated by a subscript mn , signifying a transition from energy level $m \rightarrow n$, and intra-band dephasing processes are described by a subscript mm , characterizing losses due to a change in momentum.

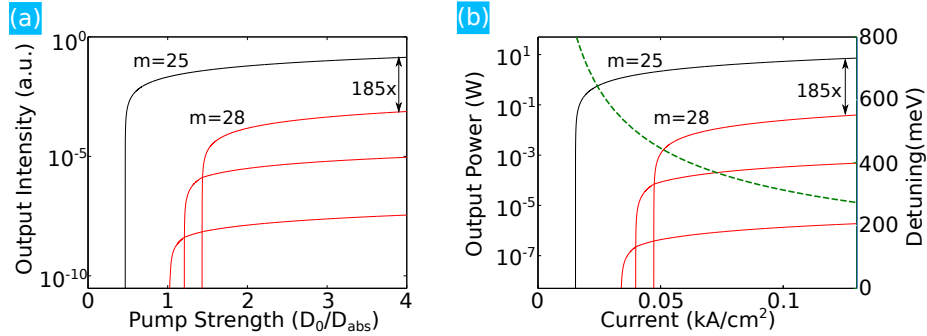


FIG. S3. (a) Reproduced from the manuscript, this figure shows the output power in the far-field as calculated for the selectively pumped cavity (solid black line), and the three strongest modes of the uniformly pumped cavity (solid red lines). (b) The same calculation as (a) but for the QCL gain medium described in the text. Power output, shown as a function of current and detuning, is in agreement with (a) up to a re-scaling determined by the reparametrization.

In the steady-state, we obtain the following analytic expression for the voltage-dependent detuning between the u and i levels,

$$\delta_{iu} = \sqrt{\frac{(\Gamma_i + \Gamma_u + \Gamma_{ul})\Omega_c^2(\Gamma_{li} - \Gamma_{ul})\zeta}{\Gamma_{li}\Gamma_{ul}D_0} - \frac{(\Gamma_u + \Gamma_i + \Gamma_{ul})\Omega_c^2(2\Gamma_{li} + \Gamma_{ul})}{\Gamma_{li}\Gamma_{ul}} - \frac{(\Gamma_u + \Gamma_i + \Gamma_{ul})^2}{4}} \quad (\text{S43})$$

Here, ζ is the density of three-level systems, and $\Omega_c = g_t/\hbar$ is the coherent coupling strength between the i and u levels. With this expression, we have parametrized the detuning in terms of the inversion density D_0 , providing a direct connection between our computational parameter and the applied bias. This approach can be easily extended to obtain expressions for population levels, and hence current, all in terms of the decay parameters, inversion

density, and the coherent coupling strength. We calculate these expressions for the disk cavity simulated in the manuscript and assume a QCL gain medium with 30 stages, disk radius of $30\ \mu\text{m}$, well width of $5\ \text{nm}$, active region depth of $1.5\ \mu\text{m}$, emission wavelength of $3\ \mu\text{m}$, and $\Gamma_i = 80 \times 10^{-3}\ \text{eV}$, $\Gamma_u = 20 \times 10^{-3}\ \text{eV}$, $\Omega_c = 5 \times 10^{-3}\ \text{eV}$, $\Gamma_{ul} = 10^{12}\ \text{s}^{-1}$, and $\Gamma_{ul} = 5 \times 10^{12}\ \text{s}^{-1}$. Note that Γ_i , Γ_u , and Ω_c are given in units of eV and should be converted to s^{-1} by multiplying with $q/\hbar$ where q is the electron charge. The results of this calculation are shown in Fig. S3(b). As expected, the power enhancement factors show complete agreement between our original calculation (Fig. S3(a)) and the same calculation repeated for a QCL gain medium. In addition, the inclusion of detuning into our model serves to quantify the power output in response to an applied bias. As shown in Fig. S3(b), the power output rises with a shrinking detuning, indicating that a maximum output (and maximum corresponding current) will be obtained for zero detuning. Above or below zero detuning, the power output will symmetrically decline as the transmission of current between the i and u levels gets suppressed.

2.2. SALT for an optically pumped N -level atomic medium

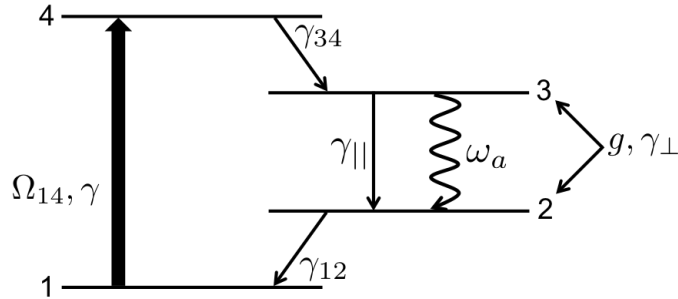


FIG. S4. The 4-level system described in the text is drawn schematically, showing all modeled decay processes and coupling energies. The solid upward arrow represents the pump from level 1 to 4 and the smaller downward arrows represent relaxation processes. The squiggly arrow indicates the lasing transition between levels 1 and 3.

While the theoretical development of SALT focused initially on a two-level system description of gain media, it is generalizable to gain media consisting of optically pumped N -level systems. To demonstrate this, we show how a general four-level system modeled using a

density-matrix approach ultimately yields equations for inversion and polarization that are identical to the Maxwell-Bloch Equations (S1)-(S3) with *effective* coefficients that can be related to the original physical parameters of the system. The derivation below is similar to that in Refs. [6, 9]. We first write the coupled equations of motion for each element ρ_{ij} in the density-matrix of the four-level system,

$$\dot{\rho}_{11} = i\Omega_{14}(\vec{r})(\rho_{41} - \rho_{41}^*) + \gamma_{12}\rho_{22}, \quad (\text{S44})$$

$$\dot{\rho}_{22} = \frac{igE(\vec{r}, t)}{\hbar}(\rho_{32} - \rho_{32}^*) + \gamma_{\parallel}\rho_{33} - \gamma_{12}\rho_{22}, \quad (\text{S45})$$

$$\dot{\rho}_{33} = -\frac{igE(\vec{r}, t)}{\hbar}(\rho_{32} - \rho_{32}^*) + \gamma_{34}\rho_{44} - \gamma_{\parallel}\rho_{33}, \quad (\text{S46})$$

$$\dot{\rho}_{44} = -i\Omega_{14}(\vec{r})(\rho_{41} - \rho_{41}^*) - \gamma_{34}\rho_{44}, \quad (\text{S47})$$

$$\dot{\rho}_{41} = -i\omega_{14}\rho_{41} - \gamma\rho_{41} - i\Omega_{14}(\vec{r})(\rho_{44} - \rho_{11}), \quad (\text{S48})$$

$$\dot{\rho}_{32} = -i\omega_a\rho_{32} - \gamma_{\perp}\rho_{32} - \frac{igE(\vec{r}, t)}{\hbar}(\rho_{33} - \rho_{22}). \quad (\text{S49})$$

As illustrated in Fig. S4, there are two well-defined laser levels (2 and 3), and 1 and 4 are the auxiliary levels. We assume that the 'pump' is a coherent optical field $E_{14}(\vec{r})$ resonant with the $1 \rightarrow 4$ transition, with the Rabi frequency $\Omega_{14}(\vec{r}) = g_{14}E_{14}(\vec{r})$, γ is the decay rate of the polarization (i.e. coherence) between levels 1 and 4, and γ_{nm} signify population relaxation rates between levels $m \rightarrow n$. ω_a is the atomic transition frequency between the laser levels 2 and 3, $\gamma_{\perp}$ and $\gamma_{\parallel}$ are the transversal and longitudinal losses as defined in the main text, and g is the dipole moment associated with this transition. It is easy to see that Equation (S49) describes the time-evolution of the transition dipole moment and is directly comparable to Equation (S2). Similarly, an equation of motion for the laser inversion can be obtained by taking the difference of Equations (S46) and (S45) and eliminating ρ_{11} and ρ_{44} in the steady state,

$$\dot{\rho}_{33} - \dot{\rho}_{22} = -2ig\frac{E(\vec{r}, t)}{\hbar}(\rho_{32} - \rho_{32}^*) - 2\gamma_{\parallel}\rho_{33} + 2\gamma_{12}\rho_{22}. \quad (\text{S50})$$

Next, we eliminate ρ_{22} and ρ_{33} using $\text{tr}(\rho) = 1$ and the resulting equation, when multiplied by the atomic density, n_a , describes the time evolution of inversion within the rotating wave approximation,

$$\dot{D} = (2\gamma_{\parallel} + \zeta) \left(\frac{\zeta n_a}{2\gamma_{\parallel} + \zeta} - D \right) - \frac{2}{i\hbar} [E^+(P^+)^* - (E^+)^*P^+]. \quad (\text{S51})$$

From this it is easy to identify that our effective Maxwell-Bloch equations have the

following parameters

$$\tilde{\gamma}_{\parallel} = 2\gamma_{\parallel} + \zeta(\vec{r}), \quad (\text{S52})$$

$$\tilde{D}_0(\vec{r}) = \frac{\zeta(\vec{r})n_a}{2\gamma_{\parallel} + \zeta(\vec{r})}, \quad (\text{S53})$$

where $\zeta(\vec{r}) = 2(\gamma_{12} - \gamma_{\parallel})/(2 + \frac{2\gamma_{12}}{\gamma_{34}} + \frac{\gamma_{12}}{\Gamma(\vec{r})})$, $\Gamma(\vec{r}) = 2\gamma\Omega_{14}^2(\vec{r})/(\Omega_{14}^2(\vec{r}) + \gamma^2)$ is the pump rate. That is, Equation (S51) is identical to the Maxwell-Bloch equation for inversion with $\gamma_{\parallel}$ and $D_0(\vec{r})$ reparametrized as $\tilde{\gamma}_{\parallel}$ and $\tilde{D}_0(\vec{r})$. The value of $\tilde{D}_0(\vec{r})$ measures the inversion at each point in the cavity as a function of the gain and loss in the cavity and it will lie between $-n_a < \tilde{D}_0 < n_a$. The approach outlined here is used below to model a Quantum Cascade Laser (QCL) gain medium.

2.3. The stadium ARC laser

In this section, we aim to demonstrate the universality of applying selective pumping for enhancing laser output power. To this end, we study a stadium-shaped asymmetric resonant cavity (ARC) laser consisting of two half-disks of radius R sandwiching a rectangle of width a and height $2R$. Compared with the microdisk laser considered in the previous section, there are fewer extremely high- Q modes that can suppress the optimally outcoupled mode when the cavity is pumped uniformly, as we have discussed in the suppression of the $m = 28$ mode in the discussion of the disk cavity in the main text. Therefore, we expect the output increase induced by selective pumping to be less dramatic here, because even uniform pumping has an increased likelihood of selecting a relatively low- Q mode.

Similar to other ARC lasers slightly deformed from a microdisk, we find that the lowest threshold modes resemble whispering-gallery modes, such as the mode shown in Fig. S5 near $\omega_a R = 23.25$ that is based on a hexagonal periodic ray orbit. We note that a stadium has completely chaotic ray dynamics and short periodic orbits are unstable, including the hexagon and the diamond in Fig. S5. However, resonant modes localized on these unstable orbits still emerge as a result of “scarring” [10, 11]. Selective pumping can be conveniently employed to favor these modes in a stadium, as well as modes localized on stable and unstable orbits in other ARC lasers.

When pumped uniformly, the aforementioned hexagonal mode starts to lase at $D_0 = 1.02D_{\text{abs}}$, and there are five other modes of better out-coupling efficiency lasing at $D_0 = 4D_{\text{abs}}$

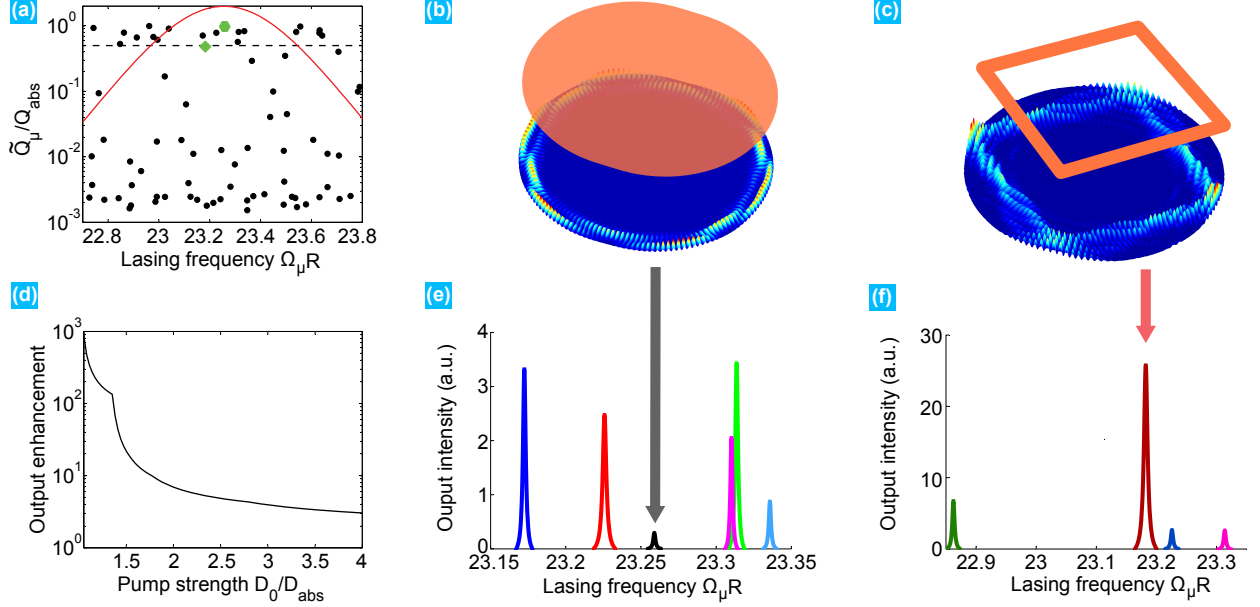


FIG. S5. (a) Total Q -factors versus the frequency for odd parity modes about the long axis in a stadium cavity of index $n_c = 3.3 + 2 \times 10^{-5}i$ and $a/R = 0.2$. Solid line shows schematically the gain curve, centered at $\omega_a R = 23.25$ and with width $\gamma_\perp R = 0.4$. Dashed line corresponds to the optimal out-coupling condition at $D_0 = 4D_{\text{abs}}$, i.e. $\tilde{Q}_\mu = Q_{\text{abs}}/2$. (b,c) False color intensity plot of the hexagonal mode with $Q_\mu \simeq 4.68 \times 10^6$ and the diamond mode with $Q_\mu \simeq 7.41 \times 10^4$ marked in (a). Shadows show the uniform and stripe-shaped pump profile. (d) Output enhancement with the stripe-shaped pump profile in (c). (e,f) Lasing spectrum at $D_0 = 4D_{\text{abs}}$, with uniform and stripe-shaped pump profiles, respectively.

(see Fig. S5(e) and Fig. S6). The one that contributes most to the output at this pump strength is an 8-bounce mode of $Q_\mu = 3.44 \times 10^5$ and $\Omega_\mu R = 23.31$. The diamond mode in Fig. S5(c) is fully suppressed in this pump range, even though its threshold in single-mode operation would be only $2.18D_{\text{abs}}$. Its out-coupling Q -factor is the closest one to $Q_{\text{abs}} = 8.25 \times 10^4$ with a well localized mode pattern, and we select it as the targeted mode using a stripe-shaped pump profile of width $0.1R$ along the diamond orbit, which covers 16.5% of the cavity area. The threshold of this diamond mode is reduced approximately by a factor of three to $0.71D_{\text{abs}}$, which becomes the first lasing mode with this tailored pump profile. Compared with the hexagonal mode and the 8-bounce mode with uniform pumping, the output power of this selectively pumped diamond mode shows an 83- and 8-fold increase

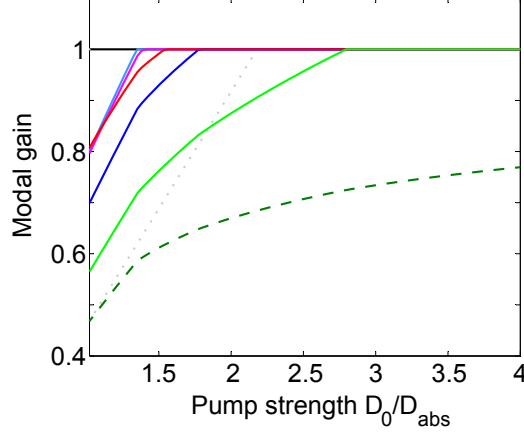


FIG. S6. Similar to Fig. 4 in the manuscript, this figure shows the modal gain for the uniformly pumped ARC cavity simulated in Fig. S5. With increasing pump, each mode, represented here by solid lines, increasingly strives towards a modal gain of unity. Upon reaching unity, each mode reaches threshold and begins to lase. There are 6 modes lasing at $D_0 = 4D_{abs}$. The dotted lines shows that the optimally outcoupled diamond mode that would have turned on at $D_0 = 2.18D_{abs}$ in the absence of modal interactions is completely suppressed in this pump range (dashed line).

at $D_0 = 4D_{abs}$, respectively, reflecting the enhancement of the out-coupling efficiency due to the tailored pump profile. Three other noticeable peaks can be seen in the output spectrum beyond $D_0 = 1.05D_{abs}$, including another diamond mode of a higher- Q value that has the second strongest output (Fig. S5(f)). If the maximum achievable pump strength is weaker and only the hexagonal mode lases with uniform pumping, e.g. at $D_0 = 1.34D_{abs}$, the output enhancement due to selective pumping can be over two orders of magnitude (Fig. S5(d)).

We note that the diamond modes and hexagonal modes discussed above can be categorized as “rotational” modes, in which light does not change its direction of rotation when propagating inside the cavity. For these modes, quasi-degeneracy occurs between $\{\sigma_x, \sigma_y\} = \{+, +\}, \{-, -\}$ symmetries and $\{+, -\}, \{-, +\}$ symmetries, respectively [10], where $\sigma_{x(y)}$ indicates the parity of the wave functions about the horizontal (vertical) axis of the cavity. Since each quasi-degenerate pair has almost identical mode patterns and out-coupling efficiencies, we consider in the main text only one from each group, namely $\{-, -\}$ and $\{-, +\}$.

2.4. The ridge cavity laser

Although modes in most laser cavities occupy spatially distinct regions, there may be some scenarios where modes are highly delocalized. The most extreme example of this is a 1D cavity such as a ridge-waveguide laser. The longitudinal modes of such a laser occupy the entire length of the cavity and consecutive modes have very high spatial overlap. Hence at first look, one may think that selective pumping should utterly fail especially in very long cavities. We show here that there is a non-trivial scheme of selective pumping that can overcome this barrier. This relies on the observation that two modes with longitudinal mode numbers n and m are periodically out of phase with a period of $2L/|n - m|$ where L is the length of the cavity. This can be seen easily by approximating any two modes in a linear cavity by $\sin(\frac{n\pi}{L}x)$ and $\sin(\frac{m\pi}{L}x)$. Then the difference of their squares yields,

$$\sin^2\left(\frac{n\pi}{L}x\right) - \sin^2\left(\frac{m\pi}{L}x\right) = \sin\left(\frac{\pi}{L}(n+m)x\right) \sin\left(\frac{\pi}{L}(n-m)x\right) \quad (\text{S54})$$

where the term on the right hand side is composed of a fast oscillating term and an envelope oscillating with the period given above. This envelope can be seen in the top figure in Fig. S7, showing the waveform of the mode that naturally lases and of the targeted mode. Our selective pumping scheme capitalizes on this behavior of modes becoming periodically out of phase to design a pumping scheme that preferentially excites the targeted mode.

We do this numerically for a cavity open on one end and use the exact modes of such a cavity. Using linear SALT theory [5], we can calculate the cavity's non-interacting thresholds and easily identify the mode with the lowest threshold under uniform pumping (labeled $m = m_{th}$ in this case). For any mode targeted for selective pumping, this low-threshold mode will be in direct competition and needs to be suppressed. Therefore, we design a contact distribution that pumps the targeted mode strongly in regions of poor overlap with the low threshold mode, and weakly (or not at all) in regions of strong overlap (see bottom figure in Fig. S7). Successful mode discrimination is achievable through this method by application of pump contacts measuring in the range of $0.15\lambda/n$ to $0.25\lambda/n$, where λ is the wavelength. Although such small contacts are not feasible in various lasers, they can be designed for THz and other long-wavelength lasers.

We numerically attempt to select the 7 modes with the lowest thresholds as determined by the linear SALT calculation, ie $m = m_{th}$ through $m = m_{th} + 6$. The pump contact used to select the 7th mode is shown in Fig. S7. Note that the contacts are only placed in selected

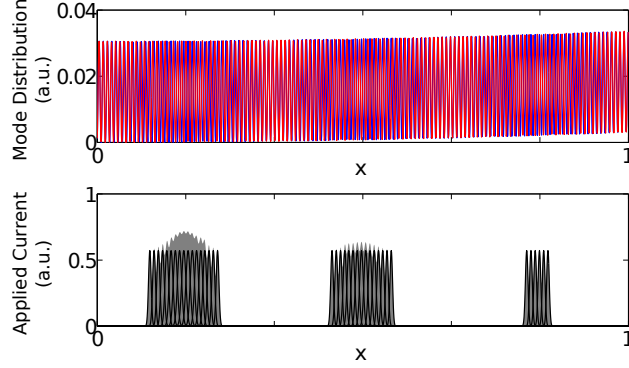


FIG. S7. In the top figure, the low-threshold mode that naturally lases under uniform pumping ($m = m_{th}$) is shown in blue and the selectively targeted mode ($m = m_{th} + 6$) is shown in red. Our pumping scheme suppresses the low-threshold mode while preferentially exciting the higher order mode by utilizing the phase relationship between the two modes as explained in the text. The pump configuration designed accordingly is shown in the bottom figure. The black gaussians represent the individual current distributions due to each applied contact, and the gray shaded regions represent the sum of the gaussians, showing the actual current distribution due to the applied contacts. The sizes and placement of the gaussians is determined by the overlap between the peaks of each mode, resulting in wider and more numerous contacts in regions of low overlap, and fewer and narrower contacts in regions of relatively higher overlap. There are no contacts in regions of very high overlap.

regions in the cavity, i.e. we do not just choose the peaks of the intensity of the mode in question. Following similar pumping schemes for each of the 7 modes, we were able to successfully achieve single-mode lasing in every case for a normalized pump power of up to $D_0/D_{abs} = 1.21$ in the best case (Fig. S8(b)). In addition, we find that the threshold of each mode is reduced slightly due to the application of selective pumping as shown in Fig. S8(a).

3. CURRENT SPREADING

Lateral current spreading is a well-known phenomenon in semiconductor lasers. In strongly diffusive media, the presence of lateral current spreading can result in a wider distribution of current at the active region than initially injected at the surface layer. In selectively pumped lasers, the proper design of pump contacts must take current spreading into account to reduce the possibility of pumping extraneous modes and reducing efficiency. To simulate

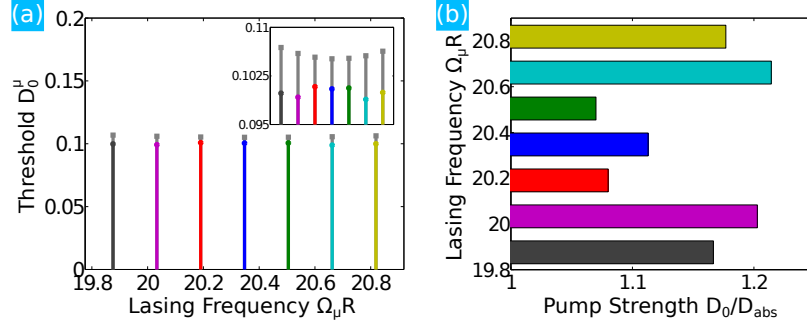


FIG. S8. (a) The lasing modes of the cavity shown in Fig. S7 are shown with their heights scaled according to their thresholds. The gray stems with square markers indicate the threshold of each cavity mode under uniform pumping. The colored stems with rounded markers are the slightly lowered thresholds due to selectively pumping each mode. The inset takes a closer look, comparing the lowered selectively pumped thresholds against the uniformly pumped case. (b) The single-mode duration of each selected mode (colors correspond to modes in (a)) in terms of pump strength is shown.

current spreading, we calculate the output power efficiency as a function of the contact width. Each contact is represented by a Gaussian at the caustic of the targeted mode with angular momentum number $m = 25$ (Fig. S9(c)). The width of each Gaussian is given by its standard deviation σ , and $\sigma = \sigma_{opt}$ is the optimal width below which the improvement in enhancement is negligible. The results for Gaussian widths ranging from $\sigma = [0.2\sigma_{opt}, 2\sigma_{opt}]$ are shown in Fig. S9(a-b).

The intensity of each mode lasing in the multimode regime can be obtained from the set of equations given in Equation (S17). From this, it is clear that the intensity of a mode already lasing will be reduced by additional modes lasing. Our calculations shown in Fig. S9(a-b) quantify this effect, showing maximum power enhancement in the single-mode regime for the selected $m = 25$ mode. Below the optimum Gaussian width, there is no discernible improvement in enhancement but above this width, a significant decline in total output power occurs due to an additional mode ($m = 27$) lasing. The relative intensities of the two lasing modes, shown in Fig. S9(b), indicate not only a sharp decline in the output of the $m = 25$ mode but a sharp decline in total emitted power. The emission from the additional mode is weak compared to the total loss in enhancement. This indicates that due to an inefficient pumping scheme, a significant amount of power is lost to regions of gain where lasing does not

take place. In addition, the extraneous $m = 27$ mode is only pumped weakly due to its poor overlap with the pump. Note also that although the pump contact width varies, the total injected power remains the same. This conservation is maintained by normalizing the pump contact distribution, resulting in a greater intensity per unit area for a narrower contact and vice versa (Fig. S9(c)).

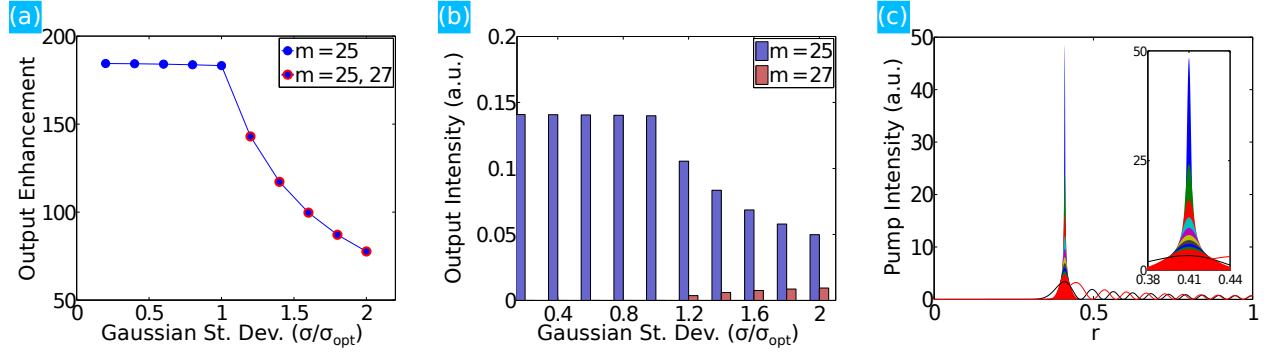


FIG. S9. (a) The output enhancement, shown for a variety of pump contacts, is maximum for $\sigma/\sigma_{opt} = 1$ and smaller. The decline for larger contacts results when mode $m = 27$ lases. (b) The output intensity distribution between modes $m = 25$ and $m = 27$. (c) The pump distributions associated with the quantities shown in (a) and (b) are plotted on top of each other, separated by color. The Bessel function waveforms illustrate the distributions of the $m = 25$ (black) and $m = 27$ (red) modes.

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