Supplementary Information

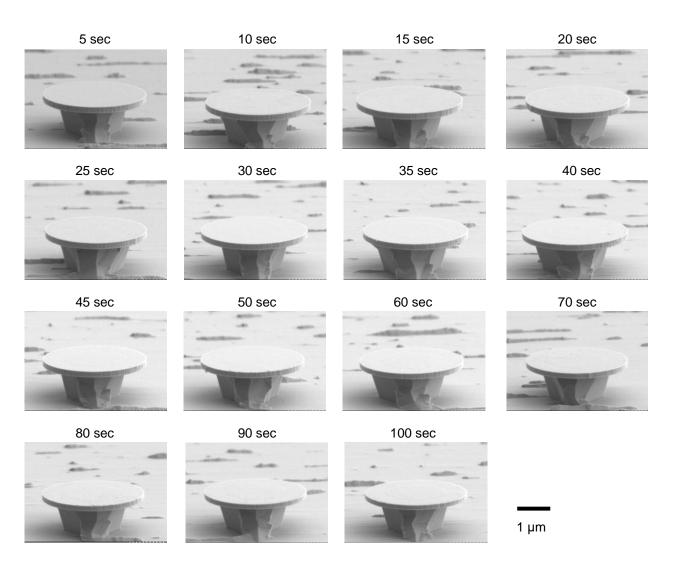
Precise photoelectrochemical tuning of semiconductor microdisk lasers

Debarghya Sarkar,^{a,b} Paul H. Dannenberg,^{a,b,c} Nicola Martino,^{a,b} Kwon-Hyeon Kim,^{a,b} Yue Wu,^{a,b} and Seok-Hyun Yun^{a,b,c*}

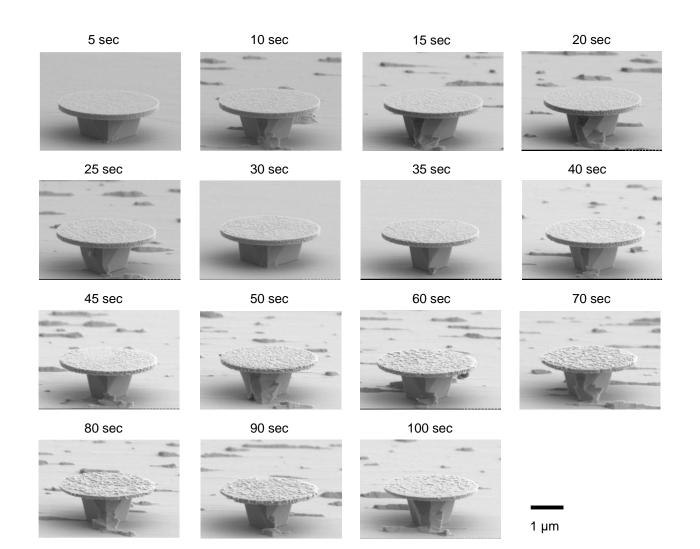
^aHarvard Medical School, Boston, Massachusetts, United States

^bMassachusetts General Hospital, Wellman Center for Photomedicine, Boston, Massachusetts, United States ^cMassachusetts Institute of Technology, Harvard-MIT Health Sciences and Technology, Cambridge, Massachusetts, United States

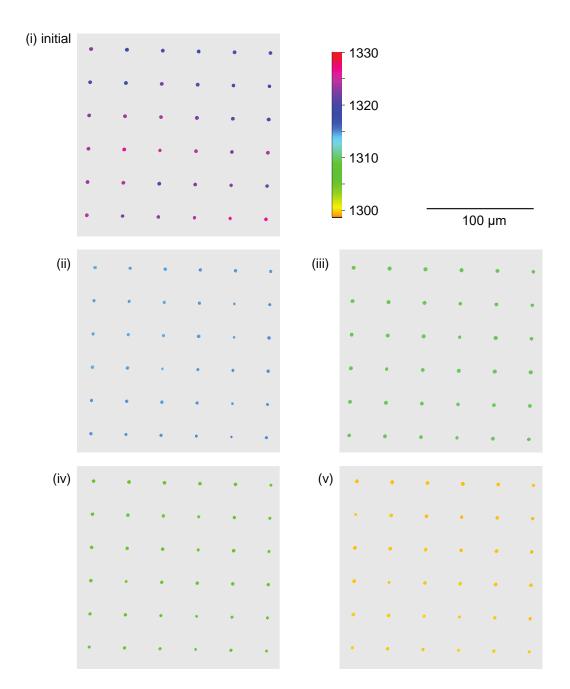
*Seok-Hyun Yun, Email: syun@hms.harvard.edu



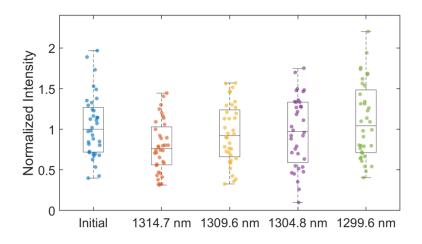
Supplementary Figure S1. SEM of silica-capped microdisk lasers with PEC tuning performed for varying times and silica cap removed thereafter. Smooth morphology with statistically reducing diameter and thickness is observed.



Supplementary Figure S2. SEM of non-silica-capped microdisk lasers with PEC tuning performed for varying times. Rough morphology with progressively worsening structure is observed.



Supplementary Figure S3. Lasing wavelength map of the disk array tuned to different narrow-band wavelengths (i) as fabricated by UV lithography and RIE, and the same sample tuned sequentially to give narrowband wavelength distributions of (ii) 1314.7 ± 0.5 nm, (iii) 1309.6 ± 0.5 nm, (iv) 1304.8 ± 0.5 nm, and (v) 1299.6 ± 0.6 nm.



Supplementary Figure S4. Intensity variation (normalized to median intensity in each case) of the disk array tuned to different narrow-band wavelengths.

Supplementary Note S1: Derivation of equilibrium band-diagram of microdisk laser in contact with etchant solution

For non-degenerately doped semiconductor, the chemical potential (μ) with respect to the energy of a free electron in vacuum is calculated as:

$$\mu = \chi + kT * \ln(N_c/N_v)$$

where χ is the electron affinity (eV), k is the Boltzmann constant (eV/K), T is the absolute temperature (K), N_c is the effective density of states in the conduction band (cm⁻³), N_v is the effective density of states in the valence band (cm⁻³).

For degenerately doped semiconductor, the chemical potential (μ) with respect to the energy of a free electron in vacuum is calculated using a more rigorous numerical analysis described in this book.³⁸

The following material parameters are used³⁴

Material	E_g (eV)	χ (eV)	$N_{c} ({\rm cm}^{-1})$	N_{v} (cm ⁻¹)
InP	1.344	4.38	5.7 x 10 ¹⁷	1.1 x 10 ¹⁹
In _{0.26} Ga _{0.74} As _{0.57} P _{0.43}	0.97	4.53	$3.5 \ge 10^{17}$	1.1 x 10 ¹⁹

Using these, the chemical potentials with respect to the vacuum energy are calculated as:

Material	Doping density (cm ⁻³)	μ (eV)
n ⁺ InP	$1 \ge 10^{18}$	4.35
UID InP	1 x 10 ¹⁶	4.48
UID In _{0.26} Ga _{0.74} As _{0.57} P _{0.43}	1 x 10 ¹⁶	4.62

Further, chemical potential of 0.5 M sulfuric acid is taken as 0.14 eV with respect to standard calomel electrode (SCE)³⁹ and therefore 4.82 eV with respect to the vacuum level.⁴⁰ For 0.0019 M sulfuric acid, assuming a chemical potential change of 54 mV/pH,³⁹ this would translate to 4.68 eV with respect to the vacuum level.

Combining these, the equilibrium band diagram of Fig. 2b is obtained. It is assumed here that, the surface of the InGaAsP and InP semiconductors in contact with water is covered with hydroxyl groups which protonate/deprotonate as the pH changes⁴¹ and thus the surface potentials of them are pH independent.

Supplementary Videos

Supplementary Video 1: Video (10x speed) of lasing wavelength tuning of an InGaAsP microdisk laser on InP pillar on InP substrate.

Supplementary Video 2: Video (2x speed) of non-tunability of lasing wavelength an InGaAsP microdisk laser released on (insulating) SiO₂ substrate.

Supplementary Video 3: Video (2x speed) of lasing wavelength tunability of InGaAsP microdisk laser released on (conducting) gold substrate.

Supplementary Video 4: Timelapse images of the uptake and physical tagging of precisely tuned laser particles by live cells in culture.