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ABSTRACT
Lasers based on Fabry–Pérot or whispering gallery resonators generally require complex fabrication stages and sensitive alignment of cavity configurations. The structural defects on reflective surfaces result in scattering and induce optical losses that can be detrimental to laser performance. On the other hand, random lasers can be simply obtained by forming disordered gain media and scatterers, but they generally show omnidirectional emission with a low Q-factor. Here, we demonstrate directional random lasers with a high Q-factor emission (∼1.5 x 10^4) via self-assembled microstructural cracks that are spontaneously formed upon radial strain-release of colloidal nanoparticles from the wet to dry phase. The rough sidewalls of cracks facilitate light oscillation via diffuse reflection that forms a spatially localized feedback, and they also serve as the laser out-coupler. These self-assembled cracks exhibit random lasing at optical pump powers as low as tens of μJ/mm². We demonstrate a wide variety of random lasers from nano- and biomaterials including silica nanoparticles, fluorescent proteins, and biopolymers. These findings pave the way toward self-assembled, configurable, and scalable random lasers for sensing, displays, and communication applications.

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INTRODUCTION
The construction of a conventional laser consists of a gain medium to amplify the light and a cavity to provide optical feedback.1 Typical cavities including Fabry–Pérot and whispering gallery resonators2 in conventional lasers trap and guide the light as it is amplified while the beam travels within the cavity boundaries by specular reflection. Since the laser emission is extremely dependent on conditions such as a high-quality surface and proper arrangement of resonator elements, the cavity design requires significant effort in such conventional lasers. Furthermore, in the case of structural defects within the cavities, resulting in light scattering or loss can be detrimental to laser performance.

In addition to conventional cavities with engineered feedback mechanisms, laser emission can also be obtained through simultaneous scattering and amplification of light in random lasers (RLs).3 The recurrent scattering creates modes as a result of coherent feedback.4 These types of lasers were first demonstrated by replacing a mirror with a scattering element in Fabry–Pérot systems, and scattering-coupled modes were achieved with uniform intensities. In RLs, omnidirectional rays due to diffuse reflection constructively interfere. So far, most of the effort on scattering based RL has been focused on spatially distributed feedback (SDF) with the use of
FIG. 1. SEM image of (a) crack patterns and (b) their rough sidewalls. (c) Schematic illustration of cracks supporting SLF based RL modes. (d) Optical microscopy image with the overlaid emission intensity of silica deposited microparticle crack pattern, with pump beam illuminating the region where lasing appears above the threshold. Laser emission is detected from bright diffuse cracks. (e) Optical setup used in RL experiments. (f) The absorption and emission spectrum of the RhB doped silica film. Emission of cracks in the spatial pattern (g) below and (h) above the threshold. (i) Emission spectrum of the cracks at different pump energy densities and (j) corresponding output energy of emission in terms of pump energy density. Inset: optical image of the pumped area below and above threshold energy, respectively.
gain media that are highly scattering. In SDF lasers, the combination of scattering points confines the beam in a closed loop and operates like a cavity.\textsuperscript{7} Advantageously, these kinds of lasers can be simply produced from a wide variety of materials such as dye-doped latex nanoparticles,\textsuperscript{8} polyhedral oligomeric silsesquioxanes,\textsuperscript{9} Ag nanoparticles embedded in polyvinyl alcohol (PVA) thin films,\textsuperscript{10} and silk inverse opals.\textsuperscript{11} Moreover, RLs have been demonstrated using biologically inspired structures such as butterfly wings,\textsuperscript{12} lotus leaves,\textsuperscript{13} biotemplated paper,\textsuperscript{14} and coral skeletons\textsuperscript{15} as they have unconventional surfaces suitable to scatter the light and emerge RL modes. Regarding chaotic structures, the scattering in RLs is omnidirectional and it is challenging to control their scattering path. Most directional RL studies mount a disordered medium to a fiber to create the waveguide effect.\textsuperscript{16} Moreover, to increase the directionality of cavities, complex nano- and micro-fabrication methods such as electron beam evaporators,\textsuperscript{17} reactive ion etching,\textsuperscript{18} and cathodic vacuum arc deposition\textsuperscript{19} were also used. Thus far, random lasers with a high Q-factor comparable to conventional lasers are fabricated, but these lasers have been shown to be omnidirectional.\textsuperscript{20} High-Q factor lasers are important in applications such as spectroscopy and metrology, but until now, distributed Bragg reflectors (DBRs) have to be integrated to disordered layers to produce directional high-Q RLs that can be used in these applications.\textsuperscript{21}

Recently, spatially localized feedback (SLF) based lasers have been established by decoupling the scattering and gain region, where the feedback is formed between two scattering layers surrounding the gain medium,\textsuperscript{22} allowing for different materials for each RL resonator element to be chosen. To date, RLs with SLF have been fabricated with pulsed laser writing via two-photon polymerization,\textsuperscript{23} laser ablation,\textsuperscript{24} and titanium dioxide nanoparticle deposition.\textsuperscript{25} Unlike previous studies, in this work, we fabricated SLF based RLs with a single step involving self-assembly. In contrast to previous crack-based random lasers,\textsuperscript{26} we demonstrate cracks oriented in the radial direction and achieve random lasers with a high Q-factor emission of $\sim 1.5 \times 10^4$ and high in-plane directionality. We made RLs from a wide variety of nano- and biomaterials including silica nanoparticles, fluorescent proteins, and biopolymers. These microlasers are created by strain release that spontaneously induces cracks with rough sidewalls. Normally, undesired crack sidewalls work well as diffusive reflectors, and output coupling occurs at the crack sidewall–air interface. Diffuse reflection from these sidewalls results in light oscillation in the pumped gain medium and lasing directionality that is quasi-orthogonal to the unparallelled crack boundaries. We demonstrate lasing in these self-assembled cavities with pump powers as low as $\sim 88 \mu J/mm^2$ for silica nanoparticles, $68 \mu J/mm^2$ for green fluorescent protein (GFP), and $38 \mu J/mm^2$ for polyvinylpyrrolidone (PVP) based cracks. Moreover, we demonstrate tuning of laser emission characteristics as regions of the microstructure with different gain spectra. These findings pave the way toward configurable and scalable integrated, self-assembled RLs for sensing,\textsuperscript{27} displays,\textsuperscript{1,28} and communication\textsuperscript{29} applications.

RESULTS AND DISCUSSION

Self-assembly of lasing microstructures

The self-assembled cracks [Fig. 1(a)] are fabricated via room-temperature drying of droplets that consist of Rhodamine B (RhB) as the gain medium, 13-nm silica nanoparticles as the crack-forming dielectric medium, and methyl ethyl ketone (MEK) and acetonitrile as the solvent. After the droplet is cast on a glass, the perimeter of the droplet pins to the glass surface. Although the solvent evaporates from the droplet surface, evaporation is faster at the perimeter where the droplet is thinnest. During the evaporation, the particles are carried to the edges by the outward radial flow of the solvent (Fig. S1 and Video S1). The faster solvent evaporation near the pinned contact line concentrates the particles close to the droplet edge. Micron-sized particles tend to form a few lines of stacked

FIG. 2. (a) Optical microscopy image with the overlaid emission intensity of RhB doped silica crack patterns when two microstructures are pumped. The pumped regions are marked with two-sided arrows. (b) Theoretically calculated mode positions for two Fabry–Pérot cavities marked with color arrows. (c) Experimentally measured emission spectrum from the region in (a).
particles at the perimeter, resulting in a well-known “coffee-ring” pattern. Nanoparticles, on the other hand, can form multilayered close-packed arrays. In the late stages of the drying process, the receding contact line induces tensile stress on the nanoparticle film, which eventually cracks to release the internal stress. While the final crack pattern depends highly on the size and softness (shear modulus) of nanoparticles, film thickness, and particle packing ratio, highly periodic and radially oriented cracks are often observed for rigid spherical nanoparticles. The drying induced cracks of nanoparticles are often considered undesirable defects; here, we use them as diffusive reflection sides [Fig. 1(b)], and the segments between the cracks function as the gain material and cavity that support SLF based RLs [Fig. 1(c)].

**Laser light generation by crack sidewalls**

To investigate the emission characteristics of self-assembled structures, the selected area of the microstructure [Fig. 1(d)] was pumped with pulses generated with an optical parametric oscillator (10 Hz, 5 ns) [Fig. 1(e)]. The pump wavelength was set to 535 nm within the absorption band of RhB for population inversion [Fig. 1(f)]. Spectral emission patterns of the cracks below and above the threshold were investigated corresponding to pump energy densities of ~40.6 \( \mu J/mm^2 \) and 253.8 \( \mu J/mm^2 \), respectively [Figs. 1(g) and 1(h)]. While the subthreshold emission pattern exhibits spectral uniformity and weak intensity, at pump power above the lasing threshold, narrower spectral lines emerge with much higher intensities. The emission spectrum of a cracked sample is illustrated at different pump energy densities [Fig. 1(i)]. The Q-factor (Q) is calculated from the ratio of the peak emission wavelength \( \lambda_{\text{Peak}} = 587 \) nm to the emission linewidth \( \Delta \lambda = 39 \) pm and is \( 1.5 \times 10^4 \). Moreover, the integrated emission intensity also shows a threshold behavior and increases significantly faster at pump powers above ~88 \( \mu J/mm^2 \) [Fig. 1(j)]. We observed linewidth reduction when the pump energy increases from sub-threshold levels (e.g., FWHM \( \approx 36 \) nm at 41.9 \( \mu J/mm^2 \)) to suprathreshold pump energy densities (e.g., FWHM \( \approx 51 \) pm at 176.8 \( \mu J/mm^2 \)). These observations support laser emission characteristics. We investigated the side facets of the cracks, and since the cracks are established by the stress relief, the side-facets are rough and scatter the light. Each microstructure creates a local optical path between the boundaries of the cracks, which is surrounded by air. In addition to their

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**FIG. 3.** (a) Fluorescence image of RhB doped silica cracks and (b) their laser emission spectrum. (c) The optical microscopy image of cracks while lasing. The green dashed lines show the pumped area. Angle marks are included. (d) The intensity of the emission profile as a function of the angle.
roughness, the consecutive side facets are also not parallel to each other. Therefore, these rough and non-parallel facets cannot support Fabry–Pérot resonances based on specular reflection. Yet, these boundaries can form SLF for the gain medium. Hence, the boundary can simultaneously lead to diffuse reflection from the scattering crack boundaries, as well as out-coupling of the generated laser light.

**Mode analysis and numerical simulations**

To explore the effect of the crack structures on the laser emission spectrum, we pumped microstructures having different widths between cracked sidewalls [Fig. 2(a)] and compared their emission profile with the calculated mode wavelengths using the $\lambda = 2n_{\text{eff}}L/m$ formula, where $\lambda$ is the wavelength, $n_{\text{eff}}$ is the effective index of the host medium, $L$ is the closed-loop path length, and $m$ is the mode number. We calculated $n_{\text{eff}} = (0.74 \times n_{\text{silica}}) + (0.26 \times n_{\text{air}}) = 1.34$, where 74% is the packing density of equal-sized spheres, the refractive index of silica $n_{\text{silica}}$ is 1.46, 26% is the fraction of air voids, and the refractive index of air $n_{\text{air}}$ is 1.0. We calculated the theoretical modes for the Fabry–Pérot cavity lengths of 60.4 $\mu$m and 73.0 $\mu$m [Fig. 2(b)], and their free spectral range (FSR) corresponds to 2.10 nm and 1.73 nm, respectively. However, the measured FSRs of 60.4 $\mu$m are 1.29 nm and 1.57 nm, and the measured FSRs of 73.0 $\mu$m cavities are 1.56 nm and 1.97 nm, which significantly differ from the calculated FSRs. Besides, we can see laser emission from

*FIG. 4. (a) The optical microscopy image with overlaid emission intensity of RhB doped silica cracks with marked laser scan direction to investigate laser wavelength emission dependence with the structure width. (b) Emission spectrum of shifted points below the threshold. The inset shows the zoomed-in view of the fitted peaks. (c) Emission spectra along the scanned line of a single microstructure that becomes narrower at greater distances. (d) Emission spectrum of shifted points shown at each 20 $\mu$m measured above the threshold. The inset shows the CMOS camera image while the cracks lase at random.*
non-parallel sidewalls on a wide variety of pumped spots on cracked microstructures, which cannot be achieved by Fabry–Pérot lasers based on specular reflection that require perfect alignment of the parallel mirrors. Moreover, the rough surface of the crack sidewalls is not appropriate for highly reflective specular reflection, indicating that the lasing can be probably supported by diffuse reflections such as the previous SLF based RLs. To sustain the phenomena that non-parallel crack side-facets constitute a RL, we examined the emission properties of the RhB doped colloidal silica droplet until it transforms into a cracked pattern. There is a time slot between solvent evaporation and crack formation. In this time interval, lasing was not observed until crack formation (see Fig. S2).

Directional random lasing

The silica crack patterns have high periodicity in the radial direction. Each zone between two consecutive cracks performs as a single microcavity since cracks are lined up next to each other with an air gap in between [Fig. 3(a)]. We pumped the radially oriented cracks using a deltoid beam with vertical symmetry to prevent the effect of the pump beam shape. The narrow peaks arose in the emission graph, indicating the SLF based random lasing [Fig. 3(b)]. The in-plane emission directionality was estimated by the far-field pattern of light intensity [Fig. 3(c)]. During laser action, emission that is orthogonal to the crack sidewalls was observed with an angle of divergence of 24° [Fig. 3(d)]. Moreover, the emission is

![Figure 5](image-url)
also partially coupled to the microcavities, which behave as optical waveguides as well.

Previously, in SDF based RLs consisting of distributed scattering points, the shift in the output spectrum was studied by adding more scattering particles to the system. In the coffee stain effect, the residue formation depends on both the particle size and shape. In another RL study, the effect of cracks on photoluminescence was analyzed and it was determined that crack-based inhomogeneous surfaces result in refractive index variations. Moreover, the spacing of particles that cause random lasing was altered by bending the substrate and a shift in RL modes was observed. In our structure, given that the structure is formed by the coffee stain effect, the gain and thickness change with the position. To map the emission features of cracks in terms of the position, we utilized an automated system to shift the sample in one direction periodically and measured the photoluminescence of cracks at each position. The optical microscopy image of the cracked pattern is shown in Fig. 4(a) where the green dashed arrow shows the direction of pump laser scan (fluorescent image of the sample is provided in Fig. S3). The spectrum images were taken along ~160 μm line in steps of ~4 μm. We illustrate the emission spectrum under the threshold pump power to check the gain medium distribution according to the position [Fig. 4(b)]. The emission spectrum blue shifts from 574.5 nm to 571.7 nm at 120 μm distance and red shifts to 572.1 nm at 140 μm. Next, we increased the pump power density above the threshold, and the position dependent output spectrum is measured along 160 μm [Fig. 4(c)]. High-intensity points with non-uniform distribution represent the random lasing based on domains between cracks. We plot the output spectra at each 20 μm in terms of wavelength extracted from Fig. 4(c), while the insets show the optical image of cracks under pump light [Fig. 4(d)]. (All measurements are represented in Video S2, and spatial emission patterns are shown in Video S3.) When we compare the gain emission spectrum with RL modes, we observe that the shift behavior is in agreement (~4 nm).

**Laser light generation by biomaterial cracks**

GFP has been widely used in biology and medicine for imaging and studying processes in cells and whole organisms. Since fluorophores of GFP are surrounded by β-barrels [Fig. 5(a)], their luminescence is more stable than other fluorescent dyes. With these protective shields, bioluminescent proteins are quite suitable optical gain materials for lasers. To date, GFP expressing cells, solid-state GFP films, or rod-shaped GFP crystals were inserted between mirrors, and their lasing properties were investigated. Furthermore, GFP is doped in silk fibroin protein and whispering gallery mode lasers, and distributed feedback lasers were built. Here, in this study, we made a GFP RL utilizing similar cracks as above. Since GFP is a bioluminescent protein, we can simultaneously use GFPs as the gain and cavity material without adding it into a host material.

To create GFP crack-based microstructures, we used aqueous GFP solution. 7 wt. % 5 μl GFP solution was dripped on a glass slide and heated inside a 50 °C oven to evaporate water, and we obtained cracked GFP stain [Fig. 5(b)]. The GFP crack formation is illustrated in Fig. S4. The absorption and emission spectra of the solidified and cracked GFP film are illustrated [Fig. 5(c)]. The measurement area for RL experiments is marked with a red, dashed circle in the white light interferometry (WLI) image of the cracked GFP layer [Fig. 5(d)]. To excite the GFP crack interfaces, we used 521 nm pump laser wavelength and a 500 nm long-pass filter. We pumped this area with different pump energy densities and observed narrow spectral peaks above ~68 μJ/mm² pump power [Figs. 5(e) and 5(f)]. Then, we separated GFP from the crack boundaries to obtain individual GFP pieces and studied their emission properties [Fig. 5(g)]. With this separation, we obtained the SLF based random lasing where the light emission is confined within the cracked sidewalls [Fig. 5(h)]. To further investigate the GFP based RLs, we examined the time-dependent intensity of the GFP crack pattern. To measure this, an ordered GFP crack zone was pumped for 40 min with 10 Hz pulsed laser at 136.5 μJ/mm² pump energy density [Fig. 5(i)]. Since the β-barrels in the GFP prevent photobleaching when the GFP is in the form of a solid film, the integrated emission field remained almost constant and the emission intensity was maintained for 40 min under 10 Hz excitation with a mean decrease less than 15%.

To motivate our method and its potential to utilize different kinds of materials, we repeated the experiments with another biopolymers. We prepared Rhodamine 6G (R6G) doped PVP crack patterns and showed that these cracks can also successfully generate SLF based random lasing. The PVP crack formation is shown in Fig. S5, while the RL experiments are represented in Fig. S6, and the lasing is observed after ~38 μJ/mm².

**CONCLUSION**

In this study, we demonstrated self-assembled, directional nano-, and biomaterial-based crack random lasing with high-Q emission. When the cavities between cracks are excited, the rough side-facets scattered the luminescence generated by fluorophores and formed optical feedback for laser light generation. First, we created silica cracks and analyzed their mode and in-plane directionality. Moreover, we showed the control of the output spectrum in terms of the spectral gain distribution. Following silica experiments, we exhibited that proteins and biopolymers can also function as RLs. We fabricated GFP and R6G doped PVP cracks and demonstrated RLs of cracks via biomaterials. In addition to the crack inside assembly, the random lasing was also observed from individual and freestanding pieces removed from the pattern through the crack boundaries. The findings pave the way toward self-assembled, configurable, and integrable RLs for sensing, displays, and communication applications.

**MATERIALS AND METHODS**

**Preparation of RhB doped silica nanoparticle colloidal solution**

13 nm silica nanoparticle-containing MEK (1 ml 30%) is doped by RhB (1 mg). Then, acetonitrile is added to the solution with 1:5 MEK to acetonitrile volume ratio.

**Preparation of GFP solution**

GFP was kindly provided by Elif Nur Firat Karalar and Deniz Conkar (Koç University, Istanbul, Turkey).
Preparation of R6G doped PVP solution

PVP (634 mg) is dissolved in water (5076 ml), and R6G (460 μg) is added to the solution.

Characterization techniques

Morphology of the cracks is analyzed with ZEISS EVO LS15 SEM operated at 3 kV. WLI measurements are performed with a Bruker Contour GT-K0 3B white light profilometer. Fluorescent properties of gain media are recorded with a steady state spectrophotometer FS5, Edinburgh Instruments. Optical microscopy images are taken using a Nikon Eclipse Ti2 microscope and an Andor Zyla 5.5 sCMOS.

Lasing spectra measurements

The wavelength of the pump laser is adjusted with an Oplet OPO, and the spectrum measurements are performed via an Andor Shamrock 500i spectrometer with 0.03 nm resolution and an Andor Newton DU970N CCD.

Image analysis

ImageJ is used to calculate the intensity density.

SUPPLEMENTARY MATERIAL

See the supplementary material for more details on the lasing data and crack fabrication.

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DATA AVAILABILITY

The data that support the findings of this study are available within the article and its supplementary material.

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