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Angular dependent light emission from planar waveguides

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We have investigated the angular dependence of amplified spontaneous emission (ASE) and laser emission from an asymmetric and free-standing polymer thin films doped with rhodamine 6G, which is transversely pumped by a pulsed Nd:YAG laser. A semi-leaky waveguide or quasi-waveguide structure has been developed by spin coating technique. In these waveguides, the light was confined by the film/air-film/glass substrate interfaces. At the film/substrate interface, a portion of light will reflect back into the film (guided mode) and the remaining refracted to the substrate resulting in cutoff modes. A blue-shift in ASE has been observed when the pump power was increased from 8 to 20 mW allowing a limited range of tuning of emission wavelength. To study the directionality of the ASE from the waveguide, we have measured the output intensity and FWHM of emission spectra as a function of viewing angle (θ) from the plane parallel to film. From the detailed examination of the output emission spectra, as $+\theta$ increases from 0° there has been an initial decrease in output intensity, but at a particular angle ≈10° an increase in output intensity was observed. This additional peak in output intensity as $+\theta$ is a clear indication of coexistence of the cutoff mode. We also present a compact solid-state laser based on leaky mode propagation from the dye-doped polymer free-standing film (∼50 μm thickness) waveguide. The partial reflections from the broad lateral surfaces of the free-standing films provided the optical feedback for the laser emission with high directionality. For a pump power of 22 mW, an intense line with FWHM <0.2 nm was observed at 578 nm. © *2015 AIP Publishing LLC*.
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INTRODUCTION

Organic luminescent materials have been widely studied in light emitting devices and solid-state lasers because of their light weight, ease of processing, low cost, flexibility, and spectral range coverage from the blue to far IR.[1] Laser devices of various structures have been achieved by a variety of resonator designs, including microcavities,[2] micro-disks,[3] micro-rings,[4] and hollow fibers.[5,6] Polymer materials have been found to be very attractive both from the technical and economical point of view with its high optical quality, better chemical compatibility with organic dyes, and inexpensive processing techniques.[7] The combination of the tunability and high efficiency of laser dyes with the high power density that can be easily achieved in waveguide structures makes devices based on dye-doped polymer waveguides and fibers very promising.[8–10]

Light amplification and spectral narrowing have been studied earlier in various systems both in the liquid and solid forms, where the spectral linewidth is reduced to a value less than 10 nm.[11–13] Spectral narrowing in most such systems is explained in terms of amplified spontaneous emission (ASE), where the spontaneously emitted luminescence is amplified by the gain medium as it propagates along the path of the maximum optical gain.[6] Reflections from the internal surfaces can increase the path length or allow multiple passes inside the gain medium which in turn build up the ASE at a faster rate.[14] Recently, laser emission from conjugated polymers and dendrimer doped polymers has also been reported.[12,15] There have been numerous investigations on laser emission from polymer planar microcavities[10,16] and polymer microring lasers.[5,17]

EXPERIMENTAL METHODS

In this section, we report a detailed study of ASE spectra observed in a thin dye-doped polymer film. To fabricate the rectangular waveguide, rhodamine 6G doped poly methyl methacrylate (PMMA) thin films with an average thickness of ∼3 μm were deposited by spin coating technique onto a glass substrate. The precursor solution had a molar composition of PMMA/Anisole/Rh6G = 2 g/15 ml/1 × 10^{-4} mol/l. Thin polymer films were formed on the glass substrate following fast drying in air. The refractive index of the doped polymer film ($n_p = 1.49$) is higher than the refractive index of the glass substrate ($n_s = 1.46$). A semi-leaky waveguide or a quasi-waveguide is obtained, where the light is confined by the film/air interface while at the film/substrate interface, a portion of light will reflect back into the film (guided mode) and the remaining refracted to the substrate (cutoff mode).[18–20] The different guided modes propagating within the gain layer have different reflectivity losses due to Fresnel law. Thus, the glass-doped polymer-air structures formed an asymmetric slab optical waveguide, which supports only the fundamental transverse mode as the guided

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mode within the emission band of the doped polymer.[1] As a result, a quasi-waveguide provides a much stronger mode restriction capability than conventional total internal reflection waveguides. However, if the incident angle is slightly smaller than the critical angle, some of the light will refract (leak) into the substrate and propagate in a direction nearly parallel to the interface between the film and substrate as shown in Fig. 1 inset.

Investigations on the ASE properties of these films were carried out by optical pumping of the films by the second harmonic output of a Q-switched Nd:YAG laser with 8 ns pulses at 10 Hz repetition rate. The experimental setup is shown in Fig. 1. The pump power was adjusted with neutral density filters and focused into the polymer-coated film by a cylindrical lens into a $1\,\text{mm} \times 0.4\,\text{mm}$ stripe. A slit was incorporated in the path of the beam between cylindrical lens and the sample so as to vary the stripe length onto the sample surface. The emission from the sample was collected by a multimode fiber and guided to a 0.5 m spectrograph coupled with a cooled CCD array. The distance between the collecting fiber and the waveguide edge was 1 cm.

RESULTS AND DISCUSSIONS

Amplified spontaneous emission

Figure 2 shows the absorption and photoluminescence spectra of the Rh6G doped PMMA film. The doped polymer film exhibits a featureless absorption band that peaks at 530 nm and emission with a peak of 570 nm, which is the characteristic emission of the Rh6G dye molecule. When the optically dense dye doped thin film is excited such that it forms a cylindrically shaped active gain medium, the fluorescence emitted by the molecules at one end is strongly amplified by the active medium and it preferentially emits the optical power along the direction of excitation, giving a highly directional output beam.[21] As a result, a large portion of the dye-doped polymer emission was optically confined within the film by internal reflections. Using the refractive index of the substrate, $n_s = 1.46$, and that of the PMMA film,

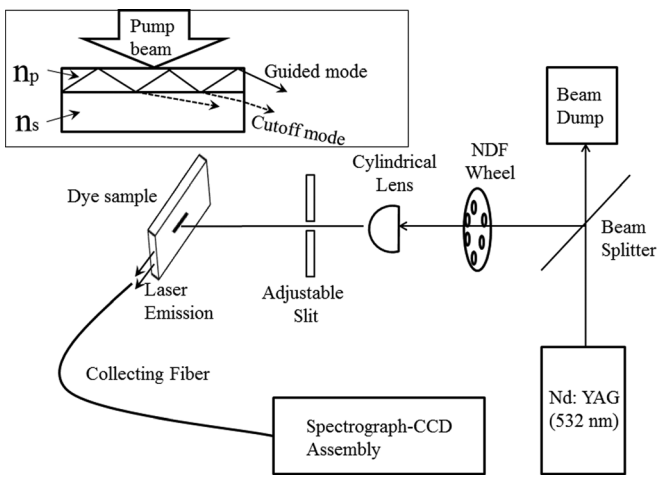

FIG. 1. Schematic representation of the experimental setup. Inset shows the Ray-tracing description of light propagating in a planar waveguide.

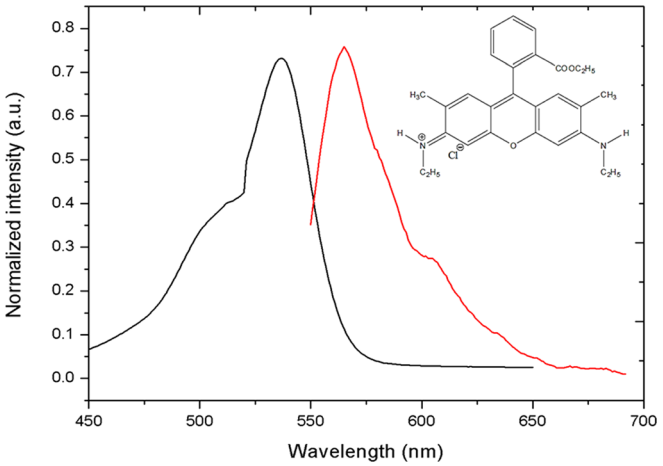

FIG. 2. The absorption and photoluminescence spectra of Rh6G doped in a PMMA film. Inset, chemical structure of Rh6G.

$n_p = 1.49$, we estimate the fraction of emission which is waveguided or "trapped" inside the film as

$$f = \sqrt{1 - \left(\frac{n_s}{n_p}\right)^2} = 0.2. \quad (1)$$

If the incident angle of the propagating light is larger than the critical angle of the interface between the film and glass substrate, the light can propagate inside the film without leakage. The spectrally narrow emission (~5 nm) from such films is observed only along the plane of the film (A in Fig. 3), whereas the emission perpendicular to the film surface remained spectrally broad (~40 nm) (B in Fig. 3). This is due to the fact that at low grazing angles, it has low loss normal to the plane of the film, and thus the polymer-air interface has almost zero transmission due to total internal reflection. This indicates that ASE is due to waveguided polymer emission, which propagates alongside the film and thus experiences the largest gain. For our measurements of ASE, we used an excitation region in the shape of a narrow stripe. As a result, ASE was predominantly emitted along the axis of stripe.

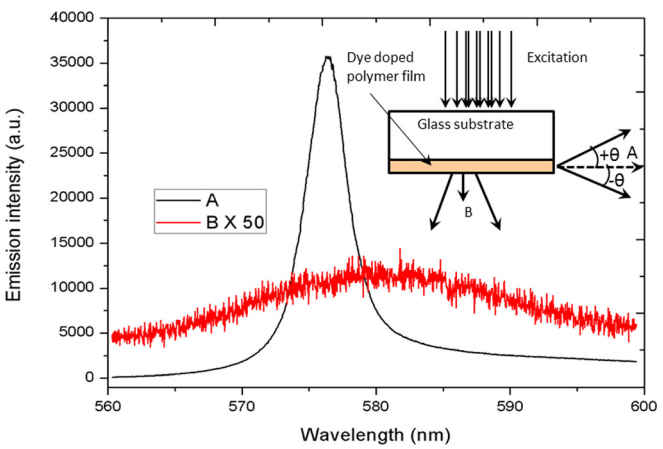

FIG. 3. The emission spectra from the dye-doped polymer film measured in the direction parallel (A) and perpendicular (B) to the film surface at the same excitation intensity (20 mW); emission intensity B is magnified by 50. The inset illustrates the experimental setup.

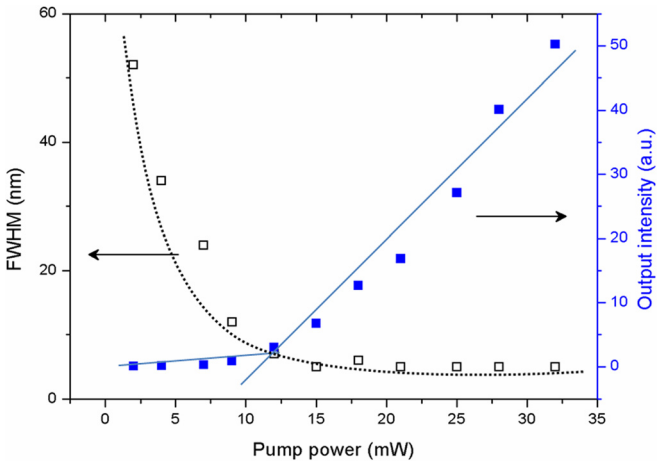

FIG. 4. Output emission intensity integrated over all wavelengths as a function of pump power (filled square) and dependence of the FWHM on the pump power (open square).

Due to stripe illumination, a large number of Rh6G dye molecules undergo population inversion when it is optically pumped by the second-harmonic output of an Nd:YAG laser. At low pump powers, the emission spectrum from the film exhibits broad fluorescence. To understand the nature of ASE in detail, the dependence of emission intensity on incident pump power is studied. As pump power is increased, the emission spectra show a line narrowing effect coupled with blue-shift as shown in Figs. 4 and 5. With an increase in pump power, the radiative transition probability gets enhanced at shorter wavelength side of the spectrum creating a shift in the spectrum towards the blue side.[5] Fig. 5 inset shows the blue-shift of the emission spectrum as a function of pump power. As the pump power increases from 8 to 20 mW, it can be seen that the emission spectrum gets shifted from 584 to 576 nm. Spectral line narrowing as a function of pump power is the signature of light amplification.[22,23] The collapses of the full-width-half-maximum (FWHM) in the emission spectrum and the change in the slope in the output emission intensity are the indication of the onset of ASE above a certain threshold ($P_{th} = 12$ mW). Also, spectral narrowing from 52 to 5 nm is observed, when the pump power is increased to 15 mW above which no further line narrowing is observed due to gain saturation effect. On a detailed study of the emission spectrum of the dye-doped polymer thin film, it is found that the emission spectrum exhibits all the features of ASE, namely, the property of directionality, narrow linewidth, and presence of soft threshold behavior.[24] Directionality of the output beam is so obvious that no focusing is required to collect the beam.

To study the directionality of the ASE from the asymmetric planar waveguide, we measure the output intensity and FWHM of emission spectra as a function of viewing angle from the plane parallel to film axis as shown in Fig. 3 inset. To measure the FWHM and output intensity as a function of viewing angle θ, the collecting fiber is kept in a rotational stage. The output intensity and FWHM of emission spectra as a function of viewing angle are measured as shown in Fig. 6. Initially, collecting fiber is aligned parallel to the waveguide axis and which is referred as angle $\theta = 0°$. Then, the FWHM and output intensity of emission spectra are measured by gradually increase/decrease angle θ ($\pm\theta$) between -40 and 40°. From the detailed examination of the output emission spectra, as $+\theta$ increases from 0° there is an initial decrease in output intensity, but at a particular angle $\approx 10°$ an increase in output intensity is observed. This additional peak in output intensity as $+\theta$ is a clear indication of coexistence of the cutoff mode.

Laser emission

In this section, we describe the observation of multimode laser emission from a transversely pumped free-standing polymer film. Solid-state free-standing thin films were prepared by incorporating Rh6G dye with PMMA. Commercially available PMMA of 5 g was dissolved in

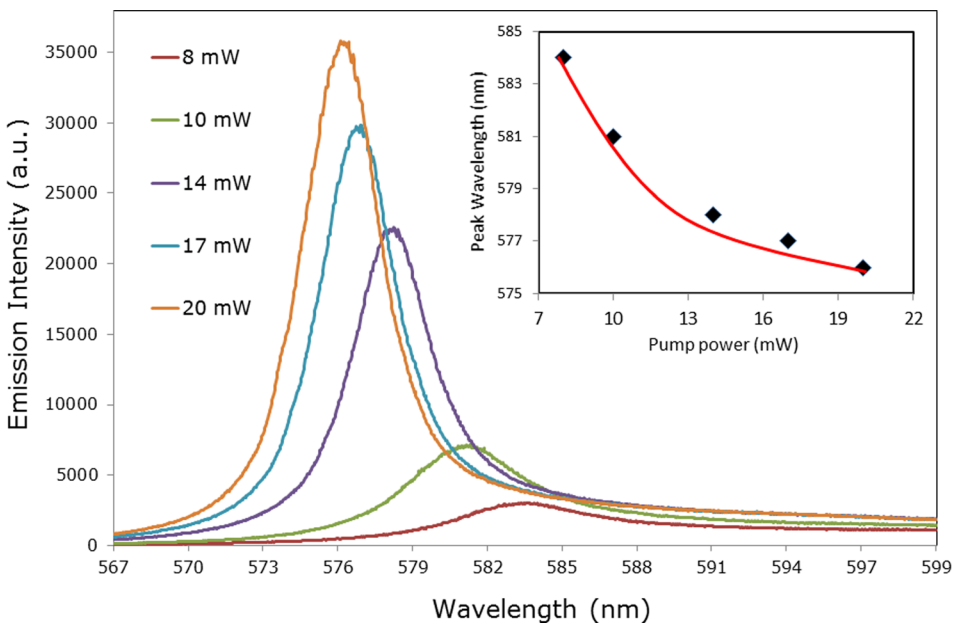

FIG. 5. The emission spectra from planar waveguide at various pump powers. Inset: Blue-shift in peak wavelength as a function of pump power.

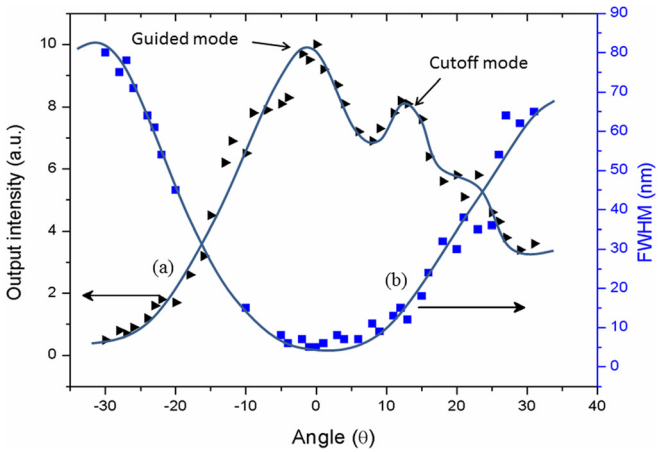

FIG. 6. The output intensity (a) and FWHM (b) of the emission spectra from planar waveguide as a function of viewing angle by keeping a constant pump power of 25 mW.

15 ml of anisole. The weight percentage was chosen to get optimum viscosity for the formation of good quality films. The dye was dissolved in this solution at a concentration of 1×10^{-4} mol/l. Films of 50 μm thickness were tape cast on glass sheets from this solution. When solvent was fully evaporated, high-quality free-standing films could be peeled off from the glass sheet. The lateral faces of the films obtained were of good optical finish and as such no further polishing was required.

The same experimental setup (explained in the Experimental Methods) was employed to study the emission characteristics from the dye-doped free-standing polymer film as well. Laser emission requires an external feedback. In the present case, no feedback is provided with external mirrors. Since the pumped polymer film was a free-standing one, the lateral faces of the film acted as mirrors, thus giving rise to a Fabry-Perot-type optical cavity for which length corresponds to the film thickness.[10] The excited active medium can be considered as a number of serially connected Fabry-Perot optical cavities. In the case of a thin stripe excitation, the stimulated emission occurs in a direction along the stripe.[25] The multiple passes between the film surfaces directly increase the gain. When the gain of the active medium compensates the losses in the medium, laser emission occurs. According to laser theory, the gain in the active medium is $\exp \sigma\,(N_2 - (g_2/g_1)N_1)l$, where σ is the stimulated emission cross section and l is the length of active medium.[24] Owing to the sample geometry and pumping scheme, the longitudinal modes of the cavity are analogous to the transverse modes of the waveguides.[10] When the pump power is increased beyond the lasing threshold (in our sample 15 mW), the emission spectrum collapsed into multiple narrow lines as shown in Fig. 7. This evenly spaced peaks clearly indicate the existence of resonant modes. Above the threshold, the total emission intensity increased much more rapidly with the pump power.

The existence of a Fabry-Perot optical cavity between the lateral faces of the free-standing film is thus confirmed by the occurrence of well-resolved discrete peaks in the emission spectrum. The mode spacing at λ can be calculated using the equation which describes Fabry-Perot cavities, namely,

$$\Delta\lambda = \frac{\lambda^2}{2nL}, \tag{2}$$

where λ is the wavelength of the strongest emission line, n is the refractive index, and L is the length of the resonator cavity. In the present case, the length of the Fabry-Perot cavity corresponds to the thickness of the polymer films. The observed mode spacing (0.9 nm) (as shown in Fig. 7) agrees well with the calculated value (0.86 nm). These observations confirm that the observed equally spaced fine structures in the emission spectra are Fabry-Perot-type modes of the optical cavity formed by the lateral faces of the waveguide.[10]

Another salient characteristic of laser emission is its increased directionality. The measurement geometry of the intensity distribution of the output from a planarmicrocavity has been shown in Fig. 7 left inset. A 1 mm pinhole was positioned in front of a collecting fiber, which was kept in a translational stage and attached to a high-speed

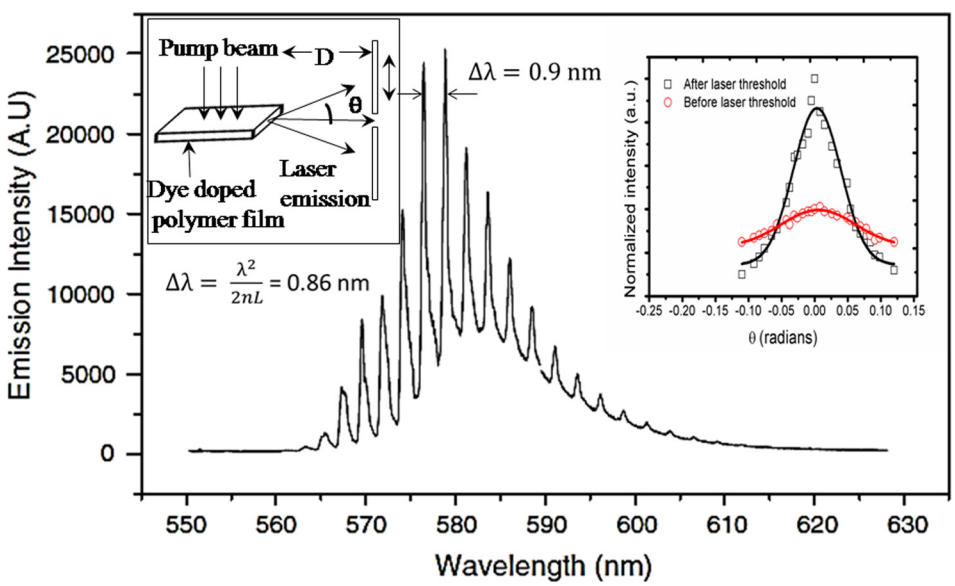

FIG. 7. Multimode laser emission from 50 μm thick rhodamine 6G doped PMMA films at a pump power of 22 mW. Left inset, schematic diagram of the setup for intensity distribution measurement. Right inset, intensity distribution of the output of a planar microcavity (data points were well-fitted with a Gaussian curve (before and after lasing threshold)).

photo-detector. The distance between collecting fiber and free-standing film was 50 mm. By varying the height of the pinhole and collecting fiber simultaneously parallel to the fiber axis, we monitored the emission intensity at a given angle θ. Below lasing threshold, the intensity distribution of emission is broad. As the pump power is increased above the lasing threshold, the lasing action occurred in the direction of the highest gain parallel to the excitation stripe ($\theta = 0°$). To confirm the spatial light confinement during the gain guiding, we measure the beam profile of the emission near the film as shown in Fig. 7 right inset. The intensity distribution is measured and the Gaussian curve is fitted on the data points from the output emission intensity above and below lasing threshold. Below lasing threshold, the regular photoluminescence emission from the planar microcavity is not highly directional, concentrated in the plane of the illuminated area of the microcavity within an angle $\Delta\theta$ of a few hundredths of a radian. Also, the emission anisotropic pattern abruptly changes above the laser threshold to $\Delta\theta = 0.1$ rad, showing excess directionality of the laser modes.

The ability of an optical resonator or cavity to confine light of a given wavelength range around λ_0 is usually measured in terms of quality factor or Q factor, which is given as

$$Q = 2\pi\nu \frac{P_{stored}}{P_{lost}} = \frac{\lambda_0}{\delta\lambda}, \quad (3)$$

where P_{stored} and P_{lost} are the power stored within the cavity and lost from the cavity, respectively, while $\delta\lambda$ is the spectral line-width of the cavity modes at wavelengths λ_0. In the present study, we estimate the Q value of the microlasers to be $>3 \times 10^3$, which results in narrow laser emission lines with a width <0.2 nm. Further improvements in the film quality may lead to enhanced directionality and high Q value of the planar microcavity laser emission.

CONCLUSION

In summary, we have presented a detailed study of the amplified spontaneous emission and laser emission from an asymmetric and free-standing planar waveguide based on a rhodamine 6G doped PMMA film. The spectrally narrow emission ($\sim$5 nm) from an asymmetric planar waveguide was observed only in the plane of the film, whereas the emission perpendicular to the film surface remained spectrally broad ($\sim$40 nm). ASE spectrum from the film exhibits high directionality, narrow linewidth, and presence of soft threshold behavior. A blue-shift in ASE has been also observed when the pump power was increased from 8 to 20 mW allowing a limited range of tuning of emission wavelength. From the study of directionality of ASE, the light amplification in an asymmetric planar waveguide is due to two different propagation modes; namely, waveguide mode and cutoff mode. Well-resolved laser modes with equal spacing were observed from a free-standing planar waveguide, which confirm the optical feedback from the Fabry-Perot-like optical cavity formed between the lateral surfaces of the thin film. The multimode laser emission from free-standing planar microcavity was characterized by narrow emission lines (<0.2 nm) with high Q ($>3 \times 10^3$) and increased directionality (0.1 rad).

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