Spectral selectivity in capillary dye lasers

ESMAIL MOBINI\textsuperscript{1,2}, BEHNA\textsuperscript{1,2}, ABAIE and ARASH MAFIF\textsuperscript{1,2,*}

\textsuperscript{1}Department of Physics & Astronomy, University of New Mexico, Albuquerque, NM 87131, USA
\textsuperscript{2}Center for High Technology Materials, University of New Mexico, Albuquerque, NM 87106, USA
\textsuperscript{*}Corresponding author: mafi@unm.edu

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We explore the spectral properties of a capillary dye laser in the highly multimode regime. Our experiments indicate that the spectral behavior of the laser does not conform with a simple Fabry-Perot analysis; rather, it is strongly dictated by a Vernier resonant mechanism involving multiple modes, which propagate with different group velocities. The laser operates over a very broad spectral range and the Vernier effect gives rise to a free spectral range which is orders of magnitude larger than that expected from a simple Fabry-Perot mechanism. The presented theoretical calculations confirm the experimental results. Propagating modes of the capillary fiber are calculated using the finite element method (FEM) and it is shown that the optical pathlengths resulting from simultaneous beatings of these modes are in close agreement with the optical pathlengths directly extracted from the Fourier Transform of the experimentally measured laser emission spectra.

\begin{align*}
\text{Eq. 1:} & \quad \frac{1}{n_{co}} + \frac{1}{n_{cl}} = \frac{1}{n_{air}} \\
\text{Eq. 2:} & \quad V = \frac{c}{2n_f} \\
\text{Eq. 3:} & \quad FSR = \frac{c}{n_f L} \\
\text{Eq. 4:} & \quad L_{eff} = \frac{1}{n_f^2} \\
\end{align*}

Dye lasers have been a major player since mid 1960s with many attractive properties including a wide operating wavelength range, often spanning 50 to 100 nanometers, and can be reasonably efficient [1, 2]. However, because of the difficulties in handling the dyes which can be poisonous or even carcinogenic, and because of their rapid degradation during operation due to photo-bleaching, dye lasers have been mostly replaced with solid state lasers. Recent advances in optofluidic systems have brought attentions back to dye lasers again [3], primarily because dyes can be recirculated in such systems in enclosed setup mitigating the disadvantages while benefiting directly from their most favorable properties. Miniaturizing liquid dye lasers into a microfluidic device has many potential advantages such as compactness, easy maintenance, safe laser operation, accurate spatial mode control, and low threshold energy [3–7] with a wide range of applications including chemical and biological sensing [8, 9]. Microfluidic fiber lasers have also appeared in optical fiber platform resulting in dye fiber lasers [10–17]. Fiber lasers based on capillary tubes and photonic crystal fibers filled with a dye solution were studied in detail by Vasdekis et al. [10]. They reported a free spectral range (FSR) which was 300 times larger than what would be expected from a simple Fabry-Perot (FP) cavity analysis: they attributed this spectral selectivity to a Vernier resonant mechanism between two transversely propagating modes in the waveguide.

In this paper, we explore the operation of a dye-filled fiber laser in the highly multi-modal regime, giving rise to multiple peaks in the laser emission spectrum, with a considerably larger spectral range (~ 50 nm) compared with the two-mode operation regime reported by Vasdekis et al. We observe that a simple FP cavity analysis does not explain the FSR for the laser spectral modes, because it predicts spectral laser lines which are much more closely spaced than what we measure in the experiment. Here, we show that a Vernier resonant mechanism involving many operating modes is responsible for the observed large value of the FSR. We develop a theoretical model which accurately predicts the FSR based on the geometrical and optical properties of the fiber and its propagating spatial modes. The calculated spectral features show a good quantitative agreement with the experiment, confirming the important role of the Vernier resonant mechanism in setting the spectral behavior of similar fiber-based systems.

The fiber dye laser used here is a fused silica capillary of 5 µm inner, and 300 µm outer diameter, respectively. Rhodamine 640 solution in benzyl alcohol at a concentration of 0.5 mg/ml was used to fill a few millimeter length of the fiber via capillary action. The benzyl alcohol solution and the cladding of the capillary have refractive indices of $n_{co} = 1.53$ and $n_{cl} = 1.46$, respectively. The relatively large refractive index contrast between fused silica cladding and benzyl solution in the fiber core enables the waveguide to operate in a highly multimodal regime. Calculations based on the $V$ number of the capillary waveguide indicate that there are nearly 70 transverse modes guided by the capillary waveguide [18]. The gain medium is pumped through an SMF-28 optical fiber to maximize the pump intensity overlap with the gain volume. The optical pump source is 0.6 ns pulse frequency-doubled Nd:YAG laser at a repetition rate of 50 Hz. The output is collected by a 40X microscope objective and is sent into a spectrometer as shown in Fig. 1.

Figures 2(a)-(d) show the emission spectra of the capillary dye laser for four different lengths of the capillary. Equation 1 is commonly used to calculated the FP cavity FSR; using this
Fig. 1. A schematic of the experimental setup. Output of a pulse frequency doubled Nd:YAG laser is coupled into a single mode fiber. The single mode fiber is butt-coupled to the dye-filled capillary. The output is collected by a 40X microscope objective and sent into a spectrometer. Pump is filtered out using a notch filter.

The equation for Fig. 2(d), e.g., gives

$$\Delta \lambda_{FSR} = \frac{\lambda^2}{2n_{cl}L} \approx 0.014 \text{ nm},$$

(1)

which is clearly much smaller than the FSR observed in Fig. 2(d).

Fig. 2. (a) Emission spectra of the capillary dye laser with an input pulse energy of $E_0$ and length of (a) $L = 3$ mm, (b) $L = 6$ mm, (c) $L = 9$ mm, and (d) $L = 11$ mm.

In order to investigate the spectral behavior of the capillary laser, we assume that there are several propagating modes undergoing multiple round trips inside the capillary. The end reflections are due to the air-fiber/solvent interfaces at the capillary tips. Ignoring the polarization dynamics and assuming linearly polarized modes, we can expand the right-moving electric field propagating inside capillary as

$$E(\rho, \varphi, z) = E_0 \sum_{l=0}^{\infty} \sum_{m=1}^{\infty} A_{lm} J_l(x_l) e^{il\varphi} e^{i\omega_{lm}z},$$

(2)

where $\beta_{lm}$ are the propagation constants of the modes. $J_l$ are Bessel functions of order $l$, which determine the transverse profile of the linearly polarized $LP_{l0}$ modes. $x_l$ is the $m$'th root of the $J_l$ Bessel function. $E_0$ is an overall constant, $A_{lm}$ is the complex amplitude coefficient of each mode, $l (l \in \{0, \sqrt{M} \})$ and $m (m \in \{1, 1, \sqrt{M} - 1/2 \})$ describe the azimuthal and radial distributions of the electric field components, respectively. $M \approx V^2/2$ is the total number of guided modes, $V = ka\sqrt{n_{co}^2 - n_{cl}^2}$ is the $V$ parameter of the waveguide, $k = 2\pi/\lambda$, and $a$ is the radius of the capillary core [18, 19].

The mode profiles and propagation constants can be numerically solved for using analytical expressions. In our analysis, we used a finite element method (FEM) code presented in Ref. [20] to calculate the propagation constants of the modes to a high accuracy.

Cavity loss from intrinsic attenuation and end mirrors are compensated by gain in the laser medium. Self-consistency requires the electric field of Eq. 2 to be reproduced after one round-trip. This translates into a series of conditions expressed as

$$e^{i\beta_{lm}2L} = 1, \quad \text{for all } l, m,$$

(3)

$$e^{i\beta_{l0}2L} = 1,$$

(4a)

$$e^{i(\beta_{lm} - \beta_{l0})2L} = 1, \quad \text{for all } l, m.$$  

(4b)

The explicit separation of the phase term corresponding to $LP_{01}$ mode makes it more convenient to apply the assumption that the $LP_{01}$ mode is dominant in power. Equation 4a results in a standard FP form expressed in Eq. 1; however, because the resulting FSR in our configuration is considerably below our spectrometer resolution, this term does not have any effect on the shape of the measured spectrum and is averaged out as observed experimentally. Rather, it is Eq. 4b as the slow oscillating spectral term, which controls the location of the spectral features in our experiment.

In order to obtain the observed FSR from Eq. 4b, let us consider the case where the phase matching is satisfied at frequency $\omega_q$, i.e., $[\beta_{01}(\omega_q) - \beta_{lm}(\omega_q)]2L = 2q\pi$, and $q$ is an integer. The FSR is determined with $q + 1$ at $\omega_{q+1}$, where a Taylor expansion results in

$$\left( \frac{\partial \beta_{01}}{\partial \omega} - \frac{\partial \beta_{lm}}{\partial \omega} \right) \delta \omega = \frac{\pi}{L}.$$

(5)

This result can be cast in terms of the group indices of the modes

$$\left( n_{01}^{(q)} - n_{lm}^{(q)} \right) \delta \omega = \frac{\pi c}{L} \rightarrow \delta \omega_{lm} = \frac{\pi c}{\Delta n_{lm}^{(q)} L}.$$

(6)
The peaks can be extracted from the Fourier transform data shown in Figs. 3(a) and (b). A simple comparison between Figs. 3(a) and (b) shows that the spacing between two neighboring peaks resulted from the 11 mm capillary is nearly two times larger than that resulted from the 6 mm capillary. This linear relationship is a strong indication of the longitudinal nature of the spectral selectivity mechanism in the capillary laser [10].

A more concrete proof can be obtained by directly calculating the values of \( d_{lm} \) using the fiber geometrical and optical parameters. Because we do not have access to the dispersion properties of the dye material, we make a reasonable assumption that the values of the group- and phase- velocity differences are nearly identical. We use the values of \( n_{co} \) and \( n_{cl} \) reported earlier and the core radius to calculate the propagation constants of the modes and establish the difference between the phase velocities of the fundamental mode and higher order modes. We note that by doing this, we are implicitly assuming that the majority of the laser power is concentrated in the fundamental mode because of the strong central pumping. It is possible to look at other mode combinations using the same formalism but we verified that our assumption captures the essence of the behavior of this capillary laser to a high degree of accuracy.

In Fig. 4, we show the values of \( d_{lm} \) directly calculated from the Fourier transform of the spectrum of the 11 mm capillary laser in Fig. 3(b): the values of \( d_{lm} \) are extracted from the positions of the major peaks in the figure and shown as black circles in Fig. 4. Using the FEM code, we calculate \( \Delta n^{(ph)} = n_{01}^{(ph)} - n_{lm}^{(ph)} \), which is the difference between the phase indices of the LP\(_{01} \) fundamental mode and the higher order modes. \( \Delta n^{(ph)} \) is substituted in Eq. 7 in place of \( \Delta n^{(g)} \), and the values of \( d_{lm} \) are shown as blue squares in Fig. 4. The strong quantitative resemblance between the experimental results and theoretical simulations indicate that our model is correct. Another way to see this result is directly in the frequency space, where e.g. the near periodic position of the peaks in Fig. 2(d) with a peak-to-peak separation of around \( \Delta \lambda \approx 5 \text{ nm} \), is almost exactly as that calculated from the beating between LP\(_{01} \) and LP\(_{11} \) modes. This agrees well with a phase refractive index difference of \( \Delta n^{(ph)} \approx 0.004 \) calculated using the FEM code. Here, we take the wavelength associated with the maximum peak in Fig. 2(d) which is around 640 nm to calculate the average FSR from Eq. 6.

![Fig. 3.](image)

**Fig. 3.** (a) Fourier transform of the emission spectrum of the 6 mm capillary dye laser. (b) Fourier transform of the emission spectrum of the 11 mm capillary dye laser.

![Fig. 4.](image)

**Fig. 4.** The black column presents the extracted optical pathlengths from Fig. 3(b). The blue column presents the resulted optical pathlengths from the calculated propagation constants inside the capillary.

Note that we only focused on the major peaks in Fig. 3(b)–minor peaks are likely related to the beating among modes, not including the LP\(_{01} \) fundamental mode. The major sources of discrepancy are likely: 1) the substitution of \( \Delta n^{(ph)} \) for \( \Delta n^{(g)} \), and 2) the beating among higher order modes which are ignored in this analysis.

In conclusion, we have reported a detailed analysis of a capillary dye laser with a highly multimodal regime of operation. The intensity spectrum of the laser shows a FSR, which is much larger than what is expected from a naive FP analysis. It is shown that the spectral selectivity is dictated by a Vernier resonant mechanism, which is predominantly driven by the beating between the LP\(_{01} \) fundamental mode and higher order modes operating in the laser. The experimental values of the FSR have a strong quantitative resemblance to those calculated using the theoretical predictions.

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