Blue amplified spontaneous emission from a stilbenoid-compound-doped polymer optical fiber

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Received September 13, 2001

We report on the fabrication and characteristics of a step-index glass-clad polymer optical fiber that uses a novel fluorescent stilbenoid compound for lasing and amplification applications. The compound, 1,4-bis(4-diphenylamino-styryl)-benzene, is specifically designed for the blue region of the spectrum and has a very high quantum yield of 0.85 in a solid-state polymer host and a large Stokes shift. Significant spectral narrowing and superlinear increase of output intensity are observed under photoexcitation at 355 nm, which are indicative of the occurrence of amplified spontaneous emission. By means of gain spectroscopy, a large optical gain of up to 36 cm^{-1} at 494 nm has been obtained for the fiber when it is transversely photoexcited at 12 mJ/cm^2. The waveguide loss has been measured to be 0.7 cm^{-1} at 494 nm. The demonstration of high gain and low waveguide loss has favorable implications for the construction of a very compact, tunable coherent light source.

OCIS codes: 060.2320, 130.0250, 140.3510, 160.2540, 250.3680, 250.5460.

The past decade has seen a surge of interest in polymer lasers, with a view to the realization of optically and electronically pumped polymer lasers.[1–3] Polymer gain media offer several advantages, such as wide spectral coverage by chemical tailoring of structure, processability that permits fabrication of devices of virtually any shape, and potentially very low cost.

Amplified spontaneous emission (ASE) and lasing characteristics of both polymers doped with small-molecular-weight compounds and conjugated polymers with a variety of device structures have been investigated. These structures include Fabry–Perot microcavities, microspheres, microcylinders, distributed Bragg reflector and distributed feedback waveguides, and photonic bandgap resonators.[4] Waveguide structures provide long gain length and optical confinement, which offer reduction of lasing threshold, a requirement for efficient lasing. Polymer fibers are especially attractive as waveguide structures because of their symmetric output-beam profile and adaptability to optical-fiber-based communication systems. There have been, however, few reports on polymer fiber lasers to date. The pioneering work by Muto *et al.* showed lasing from step-index polymer fibers with 2-cm length.[5] Kobayashi *et al.* demonstrated efficient and photostable lasing from graded-index polymer optical fibers with 5-cm length.[6] In both studies, commercially available laser dyes were used.

In this Letter, we report on the fabrication of a fluorescent-compound-doped step-index glass-clad polymer optical fiber and the observation of optical gain in the blue region. Inexpensive, compact, and tunable blue coherent light sources are particularly appealing for applications in high-density data storage as well as spectroscopy. A novel compound was designed and synthesized specifically for the blue spectral region. The inset of Fig. 1 shows the chemical structure of the compound, 1,4-bis(4-diphenylamino-styryl)-benzene (referred to as SP35). The compound has high quantum yields of 0.94 in dioxane and 0.85 in polystyrene and shows a relatively large Stokes shift of ~50 nm, which implies a low reabsorption rate, leading to ease of creating population inversion and hence to efficient light amplification. Moreover, it readily dissolves into polymer solutions. By doping of the compound at high concentration, high gain is obtainable with a short length of fiber. Henari *et al.* reported lasing characteristics of SP35 in toluene in a dye laser configuration.[7]

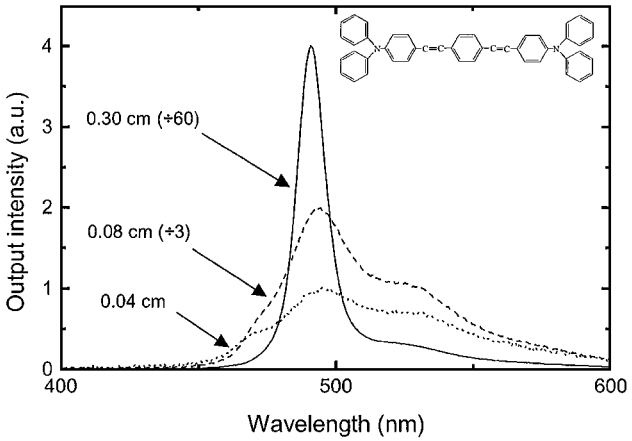

Fig. 1. Emission spectra from the fibers for excitation lengths of 0.04, 0.08, and 0.30 cm. The fluence was 12 mJ/cm^2. The inset shows the molecular structure of 1,4-bis(4-diphenylamino-styryl)-benzene (SP35).

0146-9592/01/241952-03$15.00/0

A step-index optical fiber was fabricated by use of a glass capillary as a cladding. An orthoxylene solution containing a specified amount of polystyrene and fluorescent compound was prepared. Then, the solution was drawn along the glass capillary, with a 0.5-mm inner diameter, by capillary action.

We used the stripe-excitation method to measure the optical gain in the samples. The fiber was optically excited at 355 nm by the third harmonic from a Nd:YAG laser (5-ns pulse duration, 10-Hz repetition rate). The pump beam was focused onto a 1-mm-wide stripe on the fiber by cylindrical lenses. A razor blade mounted on a translation stage was used to change the excited fiber length. The excitation fluence was kept at 12 mJ/cm^2 throughout the measurements. The output spectra from the fiber were recorded with a CCD array in conjunction with a spectrometer.

Figure 1 shows the output spectra of fibers doped with 1-wt. % SP35. The dotted curve shows the emission spectrum from the fiber with a very short excitation length of 0.04 cm. The line shape is very close to that of the spontaneous emission spectrum. The dashed curve, taken at 0.08-cm excitation length, is the emission spectrum before the onset of gain saturation. The solid curve, taken at 0.30-cm excitation length, is the almost fully narrowed emission spectrum after the onset of gain saturation. Note that the vertical scale of the excited lengths of 0.08 and 0.3 cm is reduced by 3 and 60 times, respectively. As the excitation lengths increased, the initially broad emission spectrum from the SP35-doped fiber collapsed into a narrow emission band of ~12-nm width centered at 494 nm, with an excitation length of 0.30 cm.

To illustrate further the evolution of the linewidth, we plotted the FWHM of the emission spectra as a function of the excitation length in Fig. 2. The linewidth reduces dramatically with increasing excitation length. As the excitation length is further increased, the slope gets less steep, for gain saturation sets in. The arrows in Fig. 2 indicate the data points from which the emission spectra in Fig. 1 are taken. A sudden drop in linewidth can be seen at the excitation length of ~0.10 cm. This drop occurs because, as the peak due to the 0–1 transition band grows, the lower-energy vibronic shoulder corresponding to the 0–2 transition becomes lower than the half-maximum of the 0–1 transition.

By plotting the output intensity at the peak emission wavelength versus excitation length, we can see the superlinear increase of the intensity (Fig. 3). The spectral narrowing and superlinear increase of the output intensity are indicative of ASE, also termed mirrorless lasing, resulting from amplification of spontaneously emitted light by stimulated emission as it travels through the inverted gain medium.[8]

The output intensity from one end of the fiber, $I(\lambda)$, should be given by the following expression[9]:

$$I(\lambda) = \frac{A}{\gamma(\lambda)}\{\exp[\gamma(\lambda)l] - 1\}, \qquad (1)$$

where A is a fitting parameter, $\gamma(\lambda)$ is the net gain coefficient, and l is the excited fiber length.

By carefully fitting the data to Eq. (1), we were able to obtain the net gain coefficients of the fiber. At an excitation fluence of 12 mJ/cm^2, the gain coefficient was determined to be 36 cm^{-1} at 494 nm. Since Eq. (1) is valid only for the small-signal gain region, we used the data points from 0.01 to 0.07 cm, where no gain saturation occurred, to obtain a proper net gain coefficient. The output intensity from the fiber increased exponentially with increasing excitation length, followed by gain saturation. At longer excitation lengths, the discrepancies are seen between the data and the fitted curve because of gain saturation.

Waveguide loss is also an important parameter that affects laser performance. Therefore it is imperative to assess it and to try to minimize its effects. There are two loss mechanisms: scattering and absorption. Use of fluorescent materials having a large Stokes shift can lower the loss that is due to self-absorption.

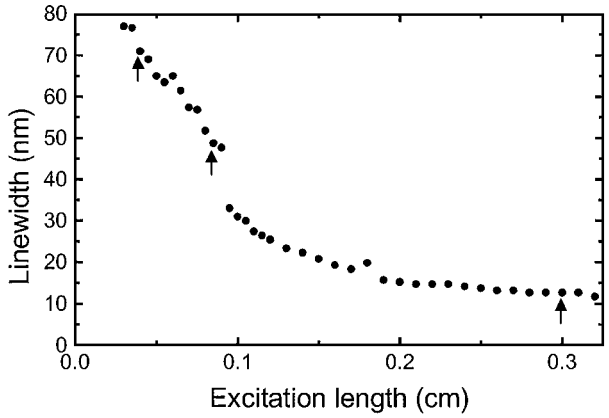

Fig. 2. Linewidth (FWHM) as a function of excitation length for the SP35-doped fiber. The fluence was 12 mJ/cm^2. The arrows indicate the data points from which the emission spectra in Fig. 1 are taken.

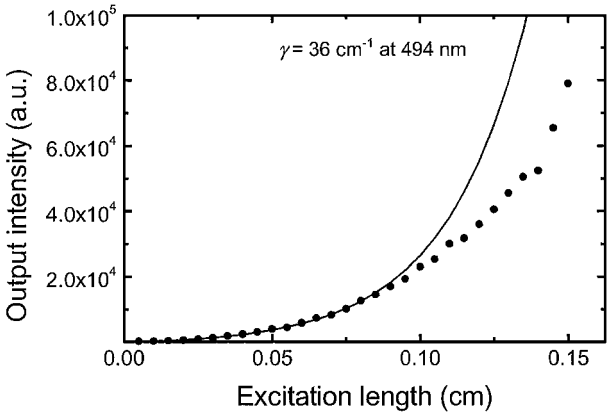

Fig. 3. Output intensity at the peak of the emission spectrum as a function of excitation length. The superlinear increase of the emission intensity can be clearly seen. The solid curve is fitted to the data by use of Eq. (1). The discrepancies between the data and the fitted curve at longer excitation length are due to gain saturation. The net gain coefficient was determined to be 36 cm^{-1} at 494 nm under photoexcitation at 12 mJ/cm^2.

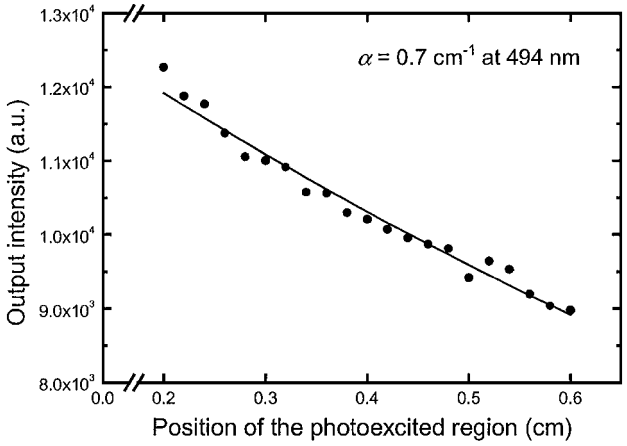

Fig. 4. Output intensity at the ASE wavelength as a function of the position of the photoexcited region. The solid curve is fitted to the data by use of the equation $I(\lambda) = I_0 \exp(-\alpha L)$, where α is a loss coefficient, L is the distance of the excited region from the fiber end, $I(\lambda)$ is the emission intensity at the fiber end, and I_0 is used as a fitting parameter. The loss coefficient was measured to be 0.7 cm^{-1} at the ASE peak wavelength of 494 nm.

Also, fluorescent materials with good solubility in host matrices do not give rise to significant scattering loss. In the present study, the waveguide loss was determined by a method similar to that for gain measurement.[10] The emission from one end of the sample was monitored when the photoexcited region was moved away from the end. Only a small region of the doped core was weakly photoexcited by use of a narrow slit with 0.1-mm width, so ASE was prevented from occurring. As the photoexcited region is moved away from the end, the emission intensity, $I(\lambda)$, coming out of the end after propagation through the unpumped region is assumed to obey the Lambert–Beer law:

$$I(\lambda) = I_0 \exp[-\alpha(\lambda)L], \tag{2}$$

where $\alpha(\lambda)$ is the loss coefficient, L is the distance of the photoexcited region from a fiber end, and I_0 is a fitting parameter. Figure 4 shows the evolution of the emission intensity at the peak ASE wavelength as a function of distance from the fiber end. By fitting the data to Eq. (2), we determined the loss to be 0.7 cm^{-1} at 494 nm. The use of the fluorescent compound with a relatively large Stokes shift and good solubility accounts for the low waveguide loss.

In summary, we have incorporated a highly fluorescent stilbenoid compound into the core region of a step-index glass-clad polymer optical fiber. On photoexcitation, the fiber exhibited optical gain in the blue region. By use of the variable-stripe method, a gain coefficient as high as 36 cm^{-1} at 494 nm has been obtained. The loss coefficient at the peak ASE wavelength has been measured to be 0.7 cm^{-1}. In the future, our work will include the fabrication of an all-solid-state polymer core fiber and demonstration of lasing. The prospects are that the high gain and low loss of the fiber presented here will allow the construction of a very compact tunable polymer fiber laser.

This work was supported by European Union ESPRIT Project 28580, A Novel Approach to Solid State Short Wavelength Laser Generation Using Luminescent Polymers (LUPO). T. Kobayashi's e-mail address is kobayast@tcd.ie.

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