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Citation: Appl. Phys. Lett. **89**, 131119 (2006); doi: 10.1063/1.2348738
View online: http://dx.doi.org/10.1063/1.2348738
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v89/i13
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Blue-green small-signal gain and saturation in a luminescent polymer gain medium

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(Received 13 February 2006; accepted 27 July 2006; published online 29 September 2006)

The authors study the optical gain and saturation behavior in a blue-green-emitting luminescent polymer gain medium. Based on the results of amplified spontaneous emission measurements, the gain coefficients, the gain-length product, and the corresponding small-signal gain are determined. By the use of the variable stripe length method, large net gain coefficients of up to 106 ± 6 cm^{-1} have been measured under nanosecond photopumping. The large gain has favorable implications for the development of short wavelength lasers and amplifiers. Their study shows that a small-signal gain of 19 dB is achievable with a very compact optical amplifier with a 400 μm length. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2348738]

Stimulated emission and lasing have been explored in a variety of polymeric gain media. These gain media include small molecule-doped polymers,[1] conjugated polymers,[2] and rare-earth-doped polymers.[3] Small-signal gain and its saturation properties are crucial for the design of an optical amplifier. No systematic study, however, has been performed on gain saturation in solid-state organic gain media. In this letter we follow the approach taken by Vehse *et al.*[4] and characterize the net optical gain and its gain saturation, the gain-length product, and the corresponding small-signal gain in a blue-green luminescent polymer gain medium.

We chose 1,4-bis[2-(4-[*N*,*N*-di(*p*-tolyl)amino]phenyl) vinyl]benzene (commercially available as B2080 from Tokyo Chemical Industry Co., Ltd.) as the active dopant and polystyrene as the host polymer. Planar waveguides were fabricated by spin coating a nitrogen-purged toluene solution containing specified amounts of polystyrene and B2080 onto Pyrex substrates. After evaporation of the solvent, the polystyrene thin films with a thickness of approximately 400 nm contained 1 wt % of B2080 and had the refractive index of 1.58 at 633 nm. The polymer thin films showed a good optical quality and no visible scattering due to the aggregated dye molecules.[5]

We used the variable stripe length method to assess the optical gain in the thin films.[6] The third harmonic output from a neodymium:yttrium aluminum garnet laser was used for transversal photopumping of the samples. The repetition rate was 10 Hz and the pulse width was 3 ns. The excitation beam is tightly focused by two cylindrical lenses into a thin-striped shape on the samples. The polarization of the pump was parallel to the propagation direction of amplified spontaneous emission induced by the thin-stripe-shaped photoexcitation. The excitation stripe length was varied by a computer-controlled mechanical shutter. The emission from the sample edge was detected with a spectrograph and a charge-coupled device camera. All the measurements were performed in an ambient atmosphere at room temperature.

The evolution of the amplified spontaneous emission intensity $I(\nu,z)$ along the propagation direction z is described by a simple one dimensional model,[7]

$$\frac{dI(\nu,z)}{dz}=\gamma(\nu)I(\nu,z)+\frac{h\nu N_2}{\tau_r}\frac{\Omega}{4\pi}, \tag{1}$$

where $\gamma(\nu)$ is the net gain coefficient, h is Planck's constant, N_2 is the excited state population density, τ_r is the radiative decay time, and Ω is the solid angle, which accounts for a fraction of spontaneously emitted light coupled into the waveguide modes for which amplification occurs. For an excitation length L, Eq. (1) can be integrated to give

$$I(\nu,L)=\frac{h\nu N_2\Omega}{4\pi\tau_r\gamma(\nu)}\{\exp[\gamma(\nu)L]-1\}$$

$$=\frac{A}{\gamma(\nu)}\{\exp[\gamma(\nu)L]-1\}, \tag{2}$$

where A is defined as $A=h\nu N_2\Omega/4\pi\tau_r$. By using $\gamma(\nu)$ and A as fitting parameters, we carefully fit the data to Eq. (2) to deduce the small-signal gain coefficients. In order to deduce gain coefficients properly, care was taken so that only data points below the onset of saturation were included in the fit. In Fig. 1, the output intensity is plotted as a function of excitation length for a series of pump power densities. The superlinear intensity increase is observed as expected for amplified spontaneous emission. Above certain excitation lengths, we can see deviation from the fitted curves, which is indicative of gain saturation due to depletion of the excited state population by stimulated emission. The onset of saturation occurs at a shorter excitation length with increasing pump power density.

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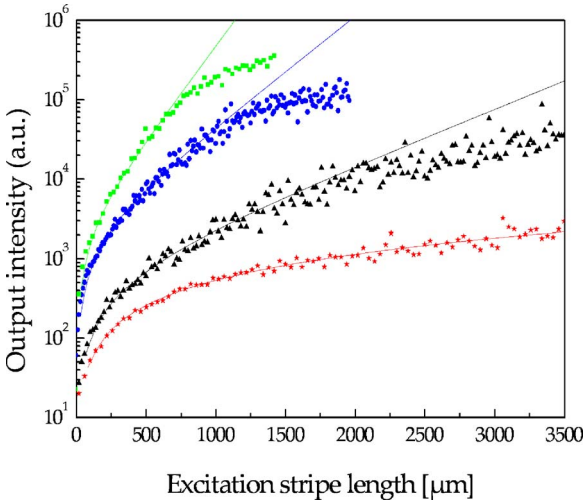

FIG. 1. (Color online) Output intensity as a function of excitation length for a series of pump power densities: 0.10, 0.25, 0.50, and 1.0 MW/cm^2 from bottom to top, respectively.

Figure 2(a) shows gain coefficients at 2.475 eV taken at different fluences. A large net gain coefficient of 106 ± 6 cm^{-1} is obtained for a pump density of 2.5 MW/cm^2 (7.5 mJ/cm^2). If we assume a simple Fabry-Pérot cavity defined by the Fresnel reflection at the polymer-air interface, we find that the maximum gain of 106 cm^{-1} is sufficient to sustain laser emission in a cavity shorter than 300 μm. This is among the largest values reported to date in organic gain media in the subnano- and nanosecond regions. Heliotis *et al.*[8] measured a net gain coefficient of 74 cm^{-1} at 2.661 eV for a pump intensity of 1.5 MW/cm^2 (0.75 mJ/cm^2) for poly(9,9-dioctylfluorene), which is one of the most extensively studied conjugated polymers operating in the short wavelength region. Our results also compare favorably with those on inorganic semiconductors. For example, a study by Frankowsky *et al.*[9] shows that, under 6 ns photopumping at 3.680 eV, a GaInN/GaN heterostructure produced a net gain of 202 cm^{-1} for 1.6 MW/cm^2 (9.6 mJ/cm^2). The magnitude of the gain exceeding 100 cm^{-1} makes our polymer gain medium attractive for the development of blue-green lasers and optical amplifiers.

The maximum small-signal gain achievable for a given pump density is a measure of amplifier performance. Figures 2(b) and 2(c) show the maximum unsaturated excitation length L_{us} and the corresponding gain-length products $\gamma(\nu)L_{us}$ for different pump power densities. From the gain-length product presented in Fig. 2(c), we calculated the small-signal gain $G(\nu)=\exp[\gamma(\nu)L_{us}]$ achievable in the polymer gain medium, as shown in Fig. 2(d). For a pump density of 2.5 MW/cm^2 (7.5 mJ/cm^2), we have a net gain of 106 ± 6 cm^{-1} and an unsaturated excitation length of 406 ± 45 μm, which results in a gain-length product of 4.3 ± 0.2, from which the maximum small-signal gain of 18.7 ± 1.0 dB is obtained.

To gain some insight into the limiting factors of the gain-length product associated with gain saturation, we consider a situation in which the output intensity is comparable to the saturation intensity. If we set $I(\nu,L)$ in Eq. (2) equal to the saturation intensity I_s and let L_s be the corresponding excitation length, the gain-length product $\gamma(\nu)L_s$ is expressed as

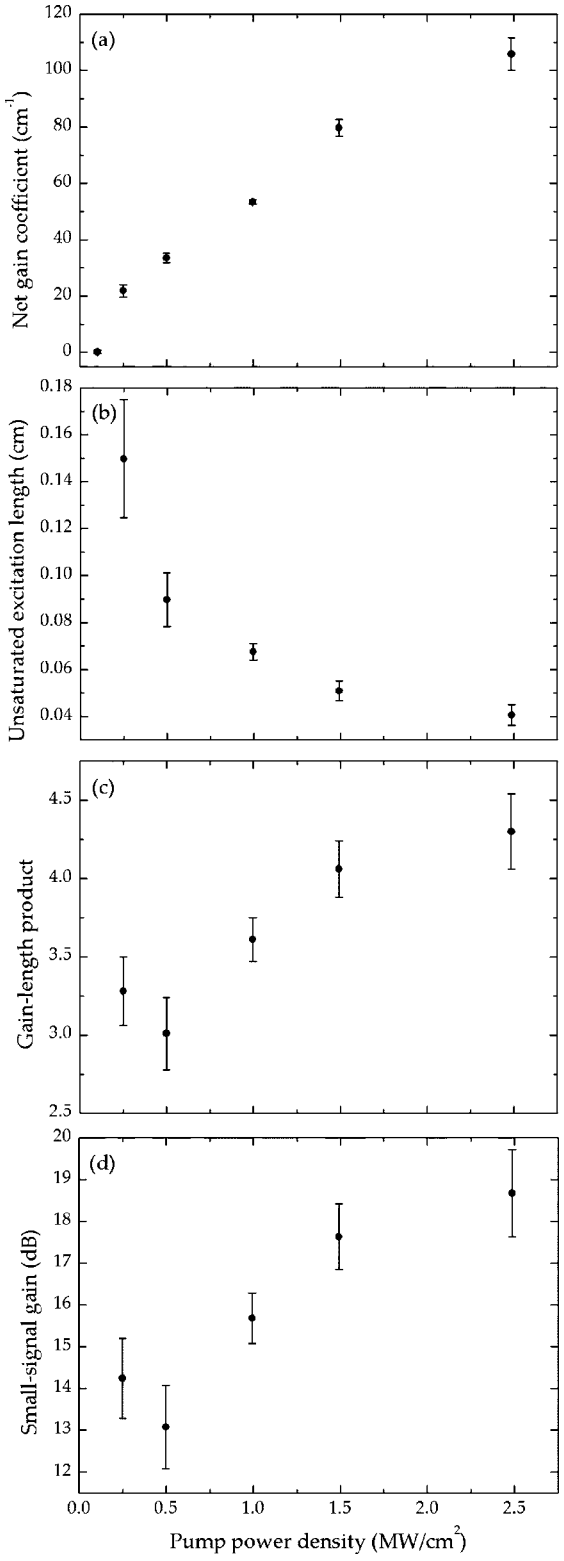

FIG. 2. (a) Net gain coefficient $\gamma(\nu)$, (b) unsaturated excitation length L_{us}, (c) gain-length product $\gamma(\nu)L_{us}$, and (d) maximum small-signal gain $G(\nu)=\exp[\gamma(\nu)L_{us}]$ plotted as functions of pump power density. For a pump density of 2.5 MW/cm^2 (7.5 mJ/cm^2), we have a net gain $\gamma(\nu)=106\pm6$ cm^{-1} and an unsaturated excitation length $L_{us}=406\pm45$ μm, which results in a gain-length product $\gamma(\nu)L_{us}=4.3\pm0.2$, from which the maximum small-signal gain $G(\nu)=\exp[\gamma(\nu)L_{us}]=18.7\pm1.0$ dB is obtained.

$$\gamma(\nu)L_s=\ln\left(1+\frac{\gamma(\nu)I_s}{A}\right). \tag{3}$$

Here $\gamma(\nu)=\sigma_e(\nu)N_2-\sigma_a(\nu)N_1-\sigma_T(\nu)N_T-\alpha$, where $\sigma_e(\nu)$, $\sigma_a(\nu)$, and $\sigma_T(\nu)$ are the transition cross sections for stimu-

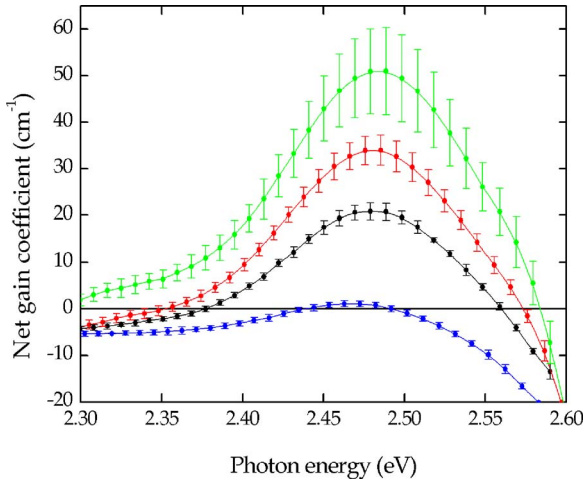

FIG. 3. (Color online) Gain spectra for a series of pump power densities: 0.10, 0.25, 0.50, and 1.0 MW/cm^2 from bottom to top, respectively. From the low energy side of the gain spectra, the waveguide loss is estimated to be $\alpha \approx 5$ cm^{-1}.

lated emission, absorption, and triplet state absorption, respectively, and α is the waveguide loss coefficient.[10] Under nanosecond photopumping, the third term on the right hand side is negligible because the pulse duration is too short to allow the population in the triplet state. As $I_s = h\nu/\sigma_e(\nu)\tau_f$, where τ_f is the fluorescent decay time, Eq. (3) becomes

$$\gamma(\nu)L_s = \ln\left[1 + \frac{\sigma_e N_2 - \sigma_a N_1 - \alpha}{\sigma_e N_2}\left(\frac{\tau_f}{\tau_r}\right)^{-1}\left(\frac{\Omega}{4\pi}\right)^{-1}\right]. \quad (4)$$

Here the ratio of the fluorescent and radiative decay times τ_f/τ_r gives the photoluminescence quantum yield of the dye molecule.[11] This simple analysis indicates that a gain medium with a lower quantum yield should give a larger gain-length product, hence a larger small-signal gain. A lower quantum yield, however, implies a larger required pump power density. Therefore there is a trade-off between the maximum small-signal gain and the required pump density. Note that the gain-length product is also a function of the solid angle Ω, which depends on the device geometry. In the present study, an asymmetric planar waveguide was used, whereas a device in a different form such as channel waveguide or optical fiber would show a different gain-length product and corresponding maximum small-signal gain.

By fitting the data to Eq. (2) at every photon energy, we were able to obtain the gain spectra for a series of pump power densities (Fig. 3). The substantial gain in the blue-green region of the spectrum has favorable implications for the development of lasers and optical amplifiers operating at around 2.4 eV, which is a potentially important communication spectral region, for some polymer waveguide materials exhibit the low loss window between 2.38 and 2.43 eV (510–520 nm).[12,13] Lastly, from the low photon energy side of the gain spectra where gain vanishes, the waveguide loss is found to be $\alpha \approx 5$ cm^{-1}.[14] This value is comparable with $\alpha = 3.5$ cm^{-1} obtained for poly(9,9-dioctylfluorene).[8]

In summary, by means of amplified spontaneous emission measurements, we have directly determined optical gain in the nanosecond regime in a polymer gain medium. Optical gains of up to 106 ± 6 cm^{-1} have been obtained in the bluecgreen region of the spectrum. Our study has shown that the polymer gain medium allows the extraction of a small-signal gain of 18.7 ± 1.0 dB in a 400-μm-long optical amplifier.

Special thanks are due to J. M. Kelly for allowing the authors to have access to the UV-visible spectrophotometer, N. Leddy, G. Armstrong, and P. Russell-Hill for the excellent technical assistance, and A. J. Fleming for stimulating discussions.

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