Continuous-wave Nd-doped polymer lasers

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Continuous-wave laser operation at 1060.2 nm was demonstrated in polymer channel waveguides doped with a Nd complex above an absorbed pump threshold of 50 mW. The highest slope efficiency of 2.15% was obtained with 5% outcoupling, resulting in a maximum output power of 0.98 mW. Lasing was also achieved on the quasi-three-level 878 nm transition above a threshold of 74.5 mW. A slope efficiency of 0.35% and an output power of 190 μm were obtained with 2.2% outcoupling. Long-term, stable cw laser operation over at least 2 h was demonstrated, indicating the durability of the polymer gain medium.

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Research in the area of solid-state polymer lasers over the past years resulted in a large number of reports on semiconducting [1] and dye-doped [2] polymer sources. Continuous-wave operation of lasers based on these gain materials is inhibited by absorption through the excited triplet state, which is characterized by a long lifetime. Because of this stringent limitation pulsed lasers are used for pumping with pulse widths ranging from 100 fs to 10 ns to ensure that the gain medium has sufficient time to recover between successive pump pulses. They typically operate at repetition rates sufficiently low (between 10 Hz and 10 kHz) to allow dissipation of the triplet populations between the pulses [1]. Although rare-earth-ion-doped polymers offer a suitable medium for generation of cw laser emission, they are notably absent from the list of polymer sources realized, which is largely due to the fluorescence quenching of the dopant ions. The latter occurs by coupling of their excited state to high-energy vibrational modes of C—H and O—H bonds both in the polymer host and in the complex, where they are encapsulated in order to be dissolved in the polymer [3], thereby imposing a requirement for high pump powers, which are usually destructive for the gain medium. Attempts to realize rare-earth polymer lasers have so far relied on pulsed pumping schemes and involved Nd^{3+}-doped chelates embedded in a poly(methyl methacrylate) (PMMA) host [4] as well as Eu^{3+}-complex-doped liquid chelate droplets [5], where heavily dumped relaxation oscillations and transient laser emission were observed, respectively.

Here, we report a cw solid-state polymer laser. The device is based on a Nd^{3+}-complex-doped polymer channel waveguide [6] and can operate on both the four-level ($^4F_{3/2} \rightarrow {}^4I_{11/2}$) and the quasi-three-level ($^4F_{3/2} \rightarrow {}^4I_{9/2}$) transitions near 1060 and 878 nm, respectively. Most important, this source does not suffer from thermal stability problems that usually plague polymer lasers and could withstand continuous laser operation for a period of at least 2 h without any signs of material degradation.

The Nd^{3+} ions were introduced into the host by encapsulation in a stable organic complex, $Nd(TTA)_3$ phen (where TTA is thenoyltrifluoro-acetone and phen is 1,10-phen anthroline), in which fluorinated chelates replaced part of the C—H bonds with C—F bonds, and the neutral ligands phen eliminated the H_2O molecules. The Nd^{3+} complex was doped into a fluorinated polymer host, 6-FDA/epoxy (where 6-FDA is 6-fluorinated-dianhydride), which has low loss around 800–1100 nm, to suppress luminescence quenching of the Nd^{3+} ions resulting from coupling of their excited state to the high-energy vibrational modes of the C—H bonds in the polymer. The chosen Nd^{3+} concentration of 1.03×10^{20} cm^{-3} is the optimal value for maximizing the attainable optical gain, recently reported to be 5.7 dB/cm around 1060 nm in channel waveguides with a cross section of 5 μm × 5 μm [7]. These channel waveguides were embedded in a cycloaliphatic-based photodefinable [8] epoxy (CHEP) cladding that was spin coated onto a thermally oxidized silicon wafer. Their fabrication involved successively photolithographic patterning of inverted channels in the CHEP layer, their backfilling with the gain medium via spin coating, thermal curing, and finally spin coating of a 3-μm-thick CHEP clad layer on top of the channels. The refractive indices of the waveguide and the CHEP cladding layer were 1.519 and 1.505 at 878 nm, and 1.516 and 1.503 at 1060 nm, respectively, as measured by using the prism coupling technique. Once fabrication was completed, the samples were cut to a length of 7.5 mm and their end faces polished to an optically smooth finish.

Laser testing of the channel waveguides was performed at room temperature by longitudinally pumping at 800 nm with a cw Ti:sapphire laser. The microscope objectives used for incoupling and outcoupling had a magnification of ×4 and ×10, respectively. The pump laser beam was expanded by using a telescope consisting of two spherical lenses with focal lengths of $f = -10$ mm and $f = 200$ mm, respectively, in order to fill the aperture of the incoupling objective. The outcoupled waveguide mode, after passing through a RG1000 or RG850 filter (for 1060 or 878 nm lasing, respectively) to block the residual pump light, was directed onto a powermeter or a spectrometer as required. Transmission measurements with the pump laser tuned off the Nd^{3+} absorption band yielded a value of 66% for the launch efficiency. The laser cavity was initially formed by attaching two thin dielectric mirrors exhibiting high reflectivity and transmission at the lasing and pump wavelength, respectively, to the end faces of the waveguide by using the surface tension of a small amount of fluorinated

liquid. Continuous-wave lasing was obtained above a threshold of 50 mW of absorbed pump power for the four-level transition near 1060.2 nm on a number of longitudinal modes as shown in the laser spectrum in Fig. 1. The wavelength separation of the peaks in the emission band corresponds to the longitudinal mode spacing $\Delta\lambda = \lambda^2/2ln = 0.049$ nm derived for a cavity length of $l = 7.5$ mm and a refractive index of $n = 1.516$ for the core material at 1060.2 nm as measured by the prism-coupling method. Measurement of the laser beam propagation factors M^2 with a Coherent Modemaster beam propagation analyzer revealed its multimode nature, yielding values of $M_x^2 = 3.25$ and $M_y^2 = 3.1$. The laser performance was further studied by using successively a set of four outcoupling mirrors with transmission values of 1.8%, 3%, 4%, and 5% at the laser wavelength. The output versus input power characteristics obtained for each of them are shown in Fig. 2. The highest slope efficiency η of 2.15% and the maximum output power of 0.98 mW for 129 mW of absorbed power were obtained by using 5% outcoupling. Lasing could not be achieved by using the outcoupling mirror with the next highest transmission available ($T = 10\%$), as the pump intensities required were destructive for the channels.

A result that exceeded our expectations was the laser operation on the quasi-three-level transition near 878 nm. Reabsorption losses combined with the relatively low stimulated-emission cross section at this wavelength ($\sigma_e \sim 1.2 \times 10^{-20}$ cm^2 as compared with $\sigma_e \sim 4.4 \times 10^{-20}$ cm^2 for the dominant four-level transition), require high pump powers, which, however, counterposes the need to maintain the pump power at a low level to avoid detrimental effects on laser performance induced by thermal degradation of the material. With two high-reflectivity mirrors used as input and output couplers, lasing was observed at an absorbed pump power threshold of 67 mW. The laser spectral output is shown in Fig. 1, where peaks correspond to the longitudinal mode spacing ($\Delta\lambda \approx 0.034$ nm) calculated from $l = 7.5$ mm and $n = 1.519$ at 878 nm. Figure 2 shows the laser output as a function of absorbed pump power for 2.2% outcoupling, which yields a slope efficiency of 0.35% and a laser threshold of 74.5 mW.

An upper limit for the propagation loss in the waveguide at 1060 nm was obtained by using the Findlay–Clay method [9]. The latter is applicable to four-level laser systems showing negligible depopulation of the ground

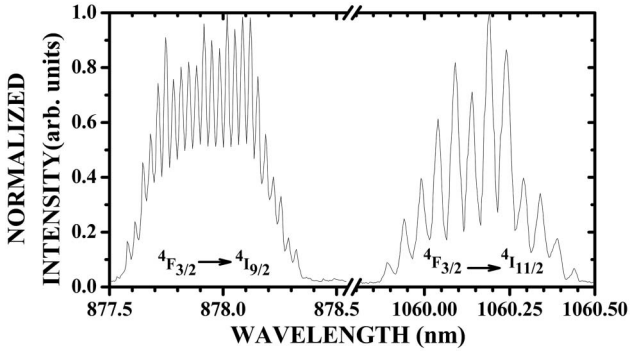

Fig. 1. Laser spectra originating from the Nd-doped polymer channel waveguides for the quasi-three-level ($^4F_{3/2} \rightarrow {}^4I_{9/2}$) and four-level ($^4F_{3/2} \rightarrow {}^4I_{11/2}$) transitions near 878.0 nm and 1060.2 nm, respectively.

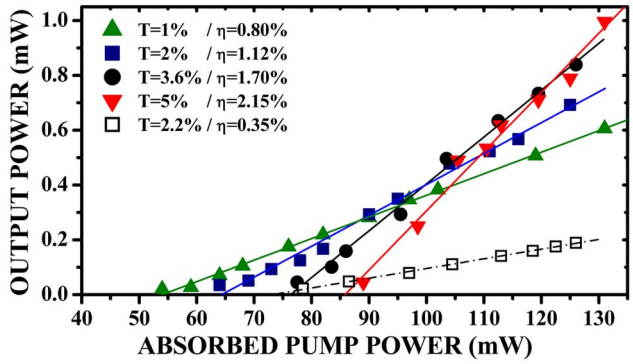

Fig. 2. (Color online) Output power as a function of absorbed pump power for the four-level and quasi-three-level laser transitions at 1060.2 nm (filled symbols) and 878.0 nm (open symbols), respectively.

state, for which the absorbed power threshold P_{th} is dependent on the outcoupling level and is given by

$$P_{\text{th}} = K[(2\alpha_L l) - \ln(R_1 R_2)], \tag{1}$$

where α_L is the propagation loss coefficient; R_1 and R_2 are the intensity reflectivities of the incoupling and outcoupling mirrors, respectively; and K is a constant dependent on the mode sizes of the laser and pump beams and material properties of the gain medium. By inserting in Eq. (1) the values for P_{th} obtained for each of the outcoupling mirrors used and then plotting $P_{\text{th}}/(2l)$ as a function of $-\ln(R_1R_2)/(2l)$, a straight line is obtained (Fig. 3). Its intercept with the abscissa yields a value of 0.032 cm^{-1} for α_L. Assuming that it is only due to propagation loss in the waveguide (in reality it also accounts for losses due to imperfections in the attachment of the cavity mirrors), it corresponds to an upper propagation loss value of 0.14 dB/cm^{-1}. While the uncertainty in the derivation of loss is rather high as a result of the low outcoupling used, the value derived is in good agreement with the propagation loss measured using the cutback method (0.1 dB/cm).

To study the consistency of the laser performance with the propagation loss derived, η was estimated by using a model that describes the effects of transverse mode profiles on η in longitudinally pumped lasers [10]. According to this theory η is given by the expression

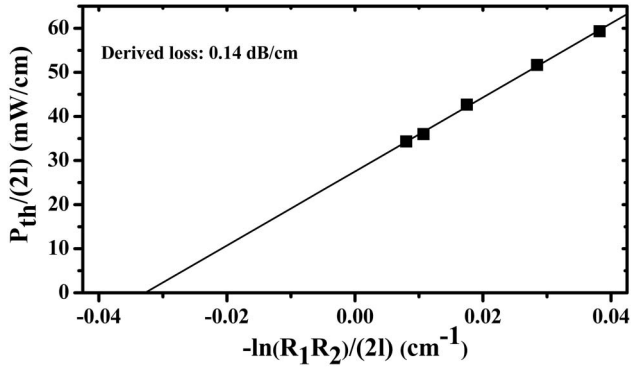

Fig. 3. Plot of $P_{\text{th}}/(2l)$ as a function of $-\ln(R_1R_2)/(2l)$. The intercept with the abscissa provides the propagation attenuation coefficient α_L.

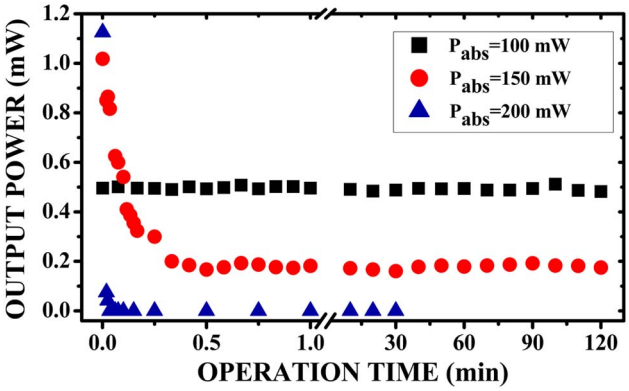

Fig. 4. (Color online) Laser output power as a function of operation time for different absorbed pump power regimes.

$$\eta = \left(\frac{\lambda_P}{\lambda_L}\right)\left(\frac{-\ln(R_2)}{-\ln(R_1R_2)+2l\alpha_L}\right)\eta_q\eta_o, \qquad (2)$$

where λ_L and λ_P are the laser and pump wavelengths, η_q is the pump quantum efficiency, and η_o is the effective overlap of the pump with the laser mode in the waveguide. Assuming unity for η_o and η_q, and $R_1 = 0.998$ and $R_2 = 0.99$, we obtain from Eq. (2) a maximum attainable value of 12.2% for η, for the specific outcoupling level. This value is significantly higher than the experimental one ($\eta = 0.8\%$) for reasons that may include a quantum efficiency substantially lower than unity, a lower-than-unity value for η_o due to the multimode nature of the waveguide, and thermal effects in the active medium, which are more severe than in crystalline waveguides.

An important aspect of the operation of optically pumped polymer lasers is their lifetime stability, which can be adversely affected by material degradation or even damage induced by the temperature increase in the active medium during pumping. The onset of such effects also determines the thermal limit for power scaling in polymer lasers. To date, the maximum reported values of lifetime stability of organic semiconductor and dye-doped lasers operating in the pulsed regime range from 10^6 to 10^7 pulses and from 10^5 to 10^6 pulses, respectively [1]. The output stability of our polymer waveguide lasers was studied as a function of operation time for three different absorbed pump power settings. In Fig. 4 it is shown conclusively that for 100 mW of absorbed pump power the active medium exhibits a level of thermal stability that is appropriate for long-term operation of at least 2 h without any signs of deterioration of the laser performance. Stable cw lasing is observed up to an absorbed pump power of 130 mW, although not all points in the input-output curves of Fig. 2 were individually tested for long-term operation over 2 h. However, for an absorbed pump power of 150 mW, the output power decreases significantly from an initial level of ~1 mW attained after the onset of pumping and stabilizes to a value of ~0.2 mW after ~20 s. This behavior is attributed to material degradation via photo-oxidation (i.e., creation of carbonyl defects in the active medium, which quench the luminescence) leading to optical loss at the pump and signal wavelengths. At absorbed powers of 200 mW, a sharp decrease in the output power is observed immediately after the onset of lasing, followed by its abrupt termination, which indicates that the temperature rise could not be sustained by the material, leading to the complete destruction of the channel.

In summary, cw laser oscillation near 1060.2 nm was demonstrated in Nd-complex-doped polymer channel waveguides with output powers of up to 0.98 mW, for pump powers low enough to allow stable laser emission for at least 2 h. Laser operation was also achieved on the quasi-three-level transition near 878.0 nm. Polymer-on-silicon is an attractive, CMOS-compatible hybrid system that exploits the ease of processing and tailoring of optical properties of organic materials. The miniature laser developed here is useful as an integrated light source for a variety of applications, ranging from light-on-chip to lab-on-chip on a silicon platform, involving IR dye and quantum-dot labeling, or as a tool for fluorescence correlation spectroscopy.

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