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Enhancement of optical gain and amplified spontaneous emission due to waveguide geometry in the conjugated polymer poly[2-methoxy-5-(2'-ethylhexyloxy)-p-phenylene vinylene]

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We report enhanced amplified spontaneous emission (ASE) and optical gain performance in a conjugated polymer (CP)-based thin film waveguide (WG) Si(100)/SiO₂/poly[2-methoxy-5-(2'-ethylhexyloxy)-p-phenylene vinylene] (MEH-PPV) by encapsulating the active layer with a transparent dielectric film of poly(methyl methacrylate) (PMMA). With index matched SiO₂ and PMMA claddings, symmetric WGs are formed that exhibit increased mode confinement and reduced propagation loss enabling lower ASE threshold (40%) and higher optical gain (50%) compared to Si(100)/SiO₂/MEH-PPV/air asymmetric WGs. An extremely large net gain coefficient of 500 cm⁻¹ is achieved under picosecond pulse excitation, which is >4× larger than values previously reported in the literature. Fabrication of symmetric WGs requires no complex processing techniques, thus offering a simple, low-cost approach for effectively controlling the ASE behavior of CP-based WGs and related optical devices. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4793422>]

Conjugated polymers (CPs) are materials that exhibit the optical and electrical properties of semiconductors while still retaining the durability and processability of plastics.¹⁻³ Conjugated polymers are also intrinsically four-level systems with large gain cross-sections making them ideal candidates for cost-effective optical components integrated on silicon. Although optically pumped Si-polymer hybrid structures have exhibited lasing action,^{4,5} achievement of stimulated emission under electrical pumping remains unsuccessful, largely because CP films cannot yet accommodate the current densities required to potentially reach threshold.⁶ Moreover, non-radiative absorption losses associated with device architecture severely degrade optical gain.⁶⁻¹⁵ While a direct injection polymer laser remains the ultimate scientific goal,¹⁶ alternative approaches are being explored such as indirect electrical pumping using, for example, light emitting diodes (LEDs)¹⁷ or pulsed microchip lasers¹⁸ as electrically driven optical pump sources for CP-based laser devices. Further optimization is required, however, and CPs exhibiting lower thresholds and higher gain are highly desirable.

We have previously shown that threshold and gain characteristics of poly[2-methoxy-5-(2'-ethylhexyloxy)-p-phenylene vinylene] (MEH-PPV) can be controlled by varying the packing morphology of the polymer chains^{19,20} and to a larger extent by optimizing the duration of the excitation pump pulses.²¹ Other approaches focus on the fabrication and design of improved device architectures. For example, Martini *et al.*²² have shown that encapsulation of MEH-PPV polymer chains in the nanopores of a silica host dramatically improves amplified spontaneous emission (ASE) threshold behavior in MEH-PPV waveguide (WG) structures. These

improvements were attributed to the nanoscale positioning and spatial alignment of the polymer chains. Threshold reductions have also been achieved by increasing confinement of the guided mode in MEH-PPV WGs formed on high-porosity porous silicon substrates.²³ Although these device architectures facilitate low threshold ASE, their fabrication is time-consuming and requires complex processing techniques. Thus, one concludes that alternative approaches must be explored if CPs are to be adopted as low-cost optical components integrated on silicon.

Here, we demonstrate significant enhancement of ASE performance in an active planar WG based on the conjugated polymer MEH-PPV. By spin-coating thin dielectric films of poly(methyl methacrylate) (PMMA) on top of Si(100)/SiO₂/MEH-PPV WGs, symmetric waveguide (SWG) heterostructures are formed that exhibit lower ASE threshold and higher net gain compared to unencapsulated Si(100)/SiO₂/MEH-PPV/air asymmetric waveguides (AWGs). Symmetric WG fabrication is low-cost and does not require complex processing or specialized equipment, thus offering an alternate route for effectively controlling the ASE characteristics of CP-based active WGs and edge-emitting distributed feedback laser devices. Note that while ASE threshold reductions have recently been demonstrated using glass/indium-tin oxide (ITO)/SiO₂/MEH-PPV/SiO₂/Ag SWG structures, the performance enhancements were achieved based on optimization of the SiO₂ spacer layer thickness; however, there was no analysis provided regarding the influence of WG symmetry and mode confinement on threshold and gain.²⁴

MEH-PPV was purchased from Sigma-Aldrich and used as received. Asymmetric WGs of Si(100)/SiO₂/MEH-PPV/air were fabricated by spin coating films of MEH-PPV from 1.0% w/v chlorobenzene solution onto Si(100) substrates with a 1-μm-thick layer of thermally grown oxide. The

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samples were then heated to 80 °C under vacuum for 60 min to remove residual solvent. Symmetric WG structures of Si(100)/SiO₂/MEH-PPV/PMMA were prepared by spin coating a layer of PMMA directly on top of the MEH-PPV films from a 5.0% w/v solution of tetrahydrofuran (THF). It has been shown that the interfacial regions between successive layers of multilayer spin-cast films are prone to enhanced degrees of interchain interactions, which is a consequence of re-dissolution of polymer chains at the surface of the previously cast film.²⁵ To minimize these effects, the PMMA cladding layer was cast from solutions containing THF as the solvent, which is well known for its ability to hinder aggregate formation.^{1,26–28} Also, the THF solvent evaporates nearly instantaneously at a spin speed of 2000 rpm, which effectively limits MEH-PPV/solvent interactions. The thicknesses of the MEH-PPV (PMMA) films, as measured by atomic force microscopy, were 120–125 nm (450–500 nm). The Si substrates were cleaved before the optical measurements to avoid nonuniformity in film thickness near the edge of the samples.

The WGs were transversely pumped using a frequency doubled ($\lambda = 532$ nm) regeneratively amplified neodymium-doped yttrium aluminum garnet (Nd:YAG) laser operating at 10 Hz. The laser pulses were 25 ps in duration and vertically polarized. The energy of the pulses was controlled using calibrated neutral density filters. Since the photoluminescence (PL) lifetime of MEH-PPV (~ 300 ps)^{29,30} is longer than the duration of the excitation pump pulses (25 ps), the measurement conditions are non-steady state. The pump beam was focused with a cylindrical lens to create a narrow excitation stripe, 100 μm wide and of variable length on the sample surface. We have verified that the collection efficiency and intensity profile were effectively constant across the lengths of stripe used. Emission was detected from the edge of the WGs using a spectrometer attached to a charge coupled device. In the subsequent discussion, the asymmetric and symmetric waveguides will be referred to as AWGs and SWGs, respectively. These samples constitute planar WGs since the refractive index of MEH-PPV ($n = 1.84$) is larger than SiO₂ ($n = 1.46$) buffer and air ($n = 1$) or PMMA ($n = 1.48$) cover layers. All of the experiments were performed under ambient conditions.

Figure 1 shows the evolution of the PL spectra for AWGs and SWGs upon increasing the pump energy density. At low pump fluence ($< 5 \mu\text{J}/\text{cm}^2$), the spectra are broad and exhibit an electronic transition at 590 nm followed by

FIG. 1. Normalized edge emitted PL spectra collected from AWGs (a) and SWGs (b) at different pump energy densities using a pump stripe length of 0.1 cm. Inset: chemical structure of MEH-PPV.

FIG. 2. Dependence of integrated emission intensity on pump energy density for AWGs (triangles) and SWGs (squares). Inset: PL linewidth (FWHM) of 0-1 transition vs. pump energy density. The pump stripe length is 0.1 cm.

vibronic progressions at 637 and 695 nm, which is characteristic of spontaneous emission. With increasing pump energy, spectral narrowing occurs at the peak of the emission spectrum dominated by the 0-1 vibronic signature at ~ 637 nm due to ASE. At sufficiently high pump energy density the broad PL spectrum collapses into a single narrowed peak with a full width at half maximum (FWHM) of ~ 10 nm (see Fig. 2 inset).

Accompanying spectral narrowing, the WGs exhibit a dramatic increase in PL efficiency above a threshold value of the pump energy density E_{th} , resulting in a sharp increase in slope as observed in Fig. 2. We define E_{th} as the pump energy density at which the PL linewidth (FWHM) of the 0-1 transition is reduced to half its initial value at low pump energy. From the FWHM vs. pump energy plot (Fig. 2 inset), the threshold values for AWGs and SWGs are $12 \mu\text{J}/\text{cm}^2$ and $7.5 \mu\text{J}/\text{cm}^2$, respectively, which means that SWGs require $\sim 40\%$ less pumping fluence to achieve threshold.

As discussed below, two contributing factors are responsible for the reduction of ASE threshold in SWGs, namely enhanced spatial confinement of the guided mode and reduced WG propagation loss.

The fundamental WG modes and associated field profiles were determined using a one-dimensional mode solver.³¹ Figure 3 shows the experimental geometry of the AWGs and SWGs with the relevant simulation parameters that were used in the WG calculations. The refractive index profile and electric field intensity distribution of the fundamental transverse electric (TE₀) guided mode for AWGs and SWGs at λ_{ASE} are shown in Fig. 4. Since transverse magnetic (TM) and higher order TE modes are not supported, only the fundamental TE₀ mode was considered in this study.

As shown in Fig. 4(a), the spatial distribution of the guided mode in the AWG exhibits an asymmetric intensity profile with a significant fraction of the mode penetrating into the SiO₂ layer. Relative to the AWG, an increase in WG

FIG. 3. Schematic of asymmetric (a) and symmetric (b) WG structures showing refractive index and layer thickness values.

FIG. 4. Refractive index profile (blue curve) and calculated electric field intensity distribution (black curve) of TE₀ guided mode for AWG (a) and SWG (b) at the ASE wavelength vs. position. Inset: Contour plot of Poynting vector (P_x) vs. position, showing density of power flowing through the WGs.

symmetry is imposed by the addition of the PMMA cover layer in the SWG which shifts the maximum field intensity of the guided mode away from the SiO₂/MEH-PPV interface towards the center of the MEH-PPV film, as observed in Fig. 4(b). Confinement of the guided mode is therefore increased^{32,33} while at the same time minimizing propagation loss, which is consistent with the reduction of the ASE threshold observed in Fig. 2.

The confinement factor Γ of a guided mode in a one dimensional planar WG structure is given by²³

$$\Gamma \approx \frac{\int_{d-\Lambda}^d S dx}{\int_{-\infty}^{\infty} S dx}, \quad (1)$$

active layer
all layers

where the x-axis is orthogonal to the plane of the WG, d is the MEH-PPV film thickness, Λ the effective absorption length ($\Lambda = \frac{1}{\alpha_{\text{abs}}} = 61$ nm, where α_{abs} is the absorption coefficient at the pump wavelength³⁴), and S the Poynting vector which describes the density of power carried by the TE₀ mode (see Fig. 4 insets). Note that since $\Lambda < d$, the optical gain initially “seen” by the confined mode is, to a first approximation, limited to the first 61 nm of the 120–125-nm-thick films. Hence, integration of the active layer is taken across the effective absorption length rather than the total film thickness. Using the aforementioned WG geometries (see Fig. 3), the confinement factors (Table I) of the TE₀ mode at λ_{ASE} for AWGs and SWGs are 0.19 and 0.25, respectively, corresponding to a 30% increase for the SWG configuration. With a larger fraction of light confined to the

TABLE I. Amplified spontaneous emission characteristics of symmetric and asymmetric WGs, where Γ is the confinement factor, α is the WG loss, E_{th} is the threshold pump fluence, and g_{max} is the maximum net gain coefficient.

WG configuration	Γ (%)	α (cm ⁻¹)	E_{th} (μJ/cm ²)	g_{max} (cm ⁻¹)
Asymmetric	19.0	11.9	12.0	331
Symmetric	24.5	8.9	7.5	501

MEH-PPV guide layer in the SWGs, more spontaneously emitted photons will be present in the stimulating field thereby increasing the probability of stimulated emission.

To characterize the effective WG losses, we performed shifting excitation stripe (SES) measurements,³⁵ which involved measuring the emission intensity I_{SES} as a function of lateral displacement of a 1 mm pump stripe away from the WG edge. The experimental data (Fig. 5) were then fitted assuming an exponential dependence on the length of the unpumped region according to $I(x, \lambda) = I_{\text{sp}} \exp[-\alpha(\lambda)x]$, where α is the loss coefficient, which includes both absorption and scattering loss, and $I_{\text{sp}}(\lambda)$ is the spontaneous emission intensity.

Loss coefficients of ~ 12 cm⁻¹ and 9 cm⁻¹ were found for AWGs and SWGs, respectively, corresponding to a 25% reduction in the total loss for symmetric WGs. This decrease in propagation loss can be explained in part by the increased confinement of guided light in the MEH-PPV gain layer. Further, the propagation angle of the TE₀ mode in the SWG is less than in the AWG, which would result in fewer side-wall reflections and hence lower scattering loss per unit length traveled by the guided light.^{36–38}

We can estimate relative changes of the pump flux ϕ_{th} at the ASE threshold for an intrinsic four-level system like MEH-PPV using the following expression for ϕ_{th} under steady-state excitation:²³

$$\phi_{\text{th}} \approx \frac{\alpha \Lambda}{\Gamma \tau \sigma_{\text{em}}}, \quad (2)$$

where τ is the excited state lifetime, σ_{em} is the stimulated emission cross-section, and the other terms as defined previously. Using the WG loss and confinement factors in Table I, a 42% reduction in the threshold is predicted for the SWG based on Eq. (2), which is in good agreement with the experimentally observed threshold reduction of 37%.

The gain performance of the WGs was measured using the variable stripe length (VSL) technique,³⁹ which involved exciting the films with a narrow pump stripe of variable length. The edge emitted peak intensity $I(\lambda)$ at λ_{ASE} was then measured as a function of the stripe length and the resulting curves fit to the expression

$$I(\lambda) = \frac{AP_o}{g(\lambda)} \left\{ e^{[g(\lambda)L]} - 1 \right\}, \quad (3)$$

where $g(\lambda)$ is the net gain coefficient, L is the pump stripe length, and AP_o describes spontaneous emission. Figure 6

FIG. 5. 0-1 peak intensity as a function of distance from sample edge for SWGs (squares) and AWGs (triangles). The solid lines are exponential fits of the data.

FIG. 6. Excitation length dependence of the peak intensity at λ_{ASE} for AWGs (a) and SWGs (b). The pump energy densities are $5 \mu\text{J}/\text{cm}^2$ (diamonds), $30 \mu\text{J}/\text{cm}^2$ (squares), and $60 \mu\text{J}/\text{cm}^2$ (triangles). Insets show the normalized PL spectra collected at three different stripe lengths with a pump energy density of $30 \mu\text{J}/\text{cm}^2$. The threshold excitation length L_t is indicated.

shows the peak intensity at λ_{ASE} for AWGs and SWGs as a function of the excitation stripe length at various pump energy densities.

Fitting of the data in Fig. 6 to Eq. (3) is given by the broken lines in the figure. Only the exponential portion of the experimental data prior to the onset of gain saturation was included in the fits. At a pump energy density of $5 \mu\text{J}/\text{cm}^2$, no amplification is present and the peak intensity grows sub-linearly with increasing stripe length. However, an exponential increase in ASE peak intensity is clearly observed for the WGs at a pump energy density of $30 \mu\text{J}/\text{cm}^2$. This exponential increase is accompanied by spectral narrowing (Fig. 6 Insets) and occurs above a threshold excitation length L_t , which decreases with increasing pump energy indicating higher gain. It should be noted that no photodamage to our samples was observed under our picoseconds pulsed excitation conditions.

Shown in Fig. 7 are the net gain coefficients as a function of the pump energy density. Each value represents the average of at least three separate measurements. At a low pump energy density of $5 \mu\text{J}/\text{cm}^2$, the net gain coefficient for the AWGs and SWGs are -10 and -8 cm^{-1} , respectively, indicating a loss. These loss values are in good agreement with the loss values obtained using the SES technique (Table I), which confirms that our experimental setup for VSL measurements is configured properly.⁴⁰

With increasing pump energy density, both the AWGs and SWGs exhibited positive net gain that scaled approximately linearly with the pump energy until eventually saturating. Extremely large gain coefficients of 330 and

FIG. 7. Net gain coefficient as a function of the pump energy density for AWGs (triangles) and SWGs (squares).

500 cm^{-1} were achieved for AWGs and SWGs, respectively, at a maximum pump energy density of $85 \mu\text{J}/\text{cm}^2$. Thus, in addition to reducing the ASE threshold, the increase in WG symmetry also serves to significantly enhance optical gain. Again, we attribute this improvement to the increased overlap between the optical WG mode and CP gain layer in the SWG relative to the asymmetric one. This is also consistent with what one would expect based on the expression for the net gain of a four-level system given by $g_{net} = g_{mod} - \alpha \approx \Gamma \sigma_{em} N_{ex} - \alpha$, where α is the loss, Γ is the confinement factor, σ_{em} is the stimulated emission cross section, g_{mod} is the modal gain, and N_{ex} is the excitation density. Total WG loss is typically much smaller than the modal gain and can therefore be neglected. Larger net gain is therefore expected from MEH-PPV WGs exhibiting larger optical confinement. Using the calculated confinement factor values (Table I), the four-level gain equation (above) predicts a factor of 1.3 increase in the maximum net gain for SWGs relative to AWGs, which is in reasonable agreement with the experimentally observed factor of 1.5. In addition to the significant enhancement of optical gain and lowered threshold for ASE that we have reported here, Richardson *et al.*⁴¹ have shown that encapsulating the MEH-PPV WG layer improves operational lifetime and stability by minimizing photooxidation. While these authors acknowledged the dependence of the waveguiding properties on the index profile of the device, the quantitative effect on optical gain was not investigated.

In summary, we have demonstrated a simple, low-cost approach for enhancement of ASE performance in MEH-PPV films using symmetric WGs with optimized distribution of the TE_0 guided mode in the CP gain layer. A low threshold of $7.5 \mu\text{J}/\text{cm}^2$ and extremely large net gain coefficient of 500 cm^{-1} were achieved in symmetric WGs under ps pulse excitation, compared to $12 \mu\text{J}/\text{cm}^2$ and 330 cm^{-1} for asymmetric WGs. These gain coefficients are a factor of 4 or higher than those previously reported in the literature. The efficacy of our methodology is attributed to enhanced optical confinement and reduced propagation loss of gain guided light in the symmetric WG structures. Fabrication of symmetric WGs requires no complex processing techniques, thus offering simpler access to improved ASE characteristics in CP-based WGs and related optical devices.

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