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Distributed feedback laser action from polymeric waveguides doped with oligo phenylene vinylene model compounds

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We report lasing studies of poly(styrene) waveguides doped with amino- and cyano-substituted oligo phenylene vinylene (distyryl benzene) model compounds under picosecond excitation. Optical feedback is provided by distributed Bragg gratings formed in the film by interference patterns in the pump beam. We demonstrate broad tunability of laser emission in these materials and simultaneous lasing at two wavelengths separated by 23 nm. Tuning ranges of the model compounds are compared with previous experiments. © *2000 American Institute of Physics.*
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We report optically induced distributed feedback (DFB) laser action studies involving two distyryl benzene derivatives: 1,4-bis-(4-diphenylamino-styryl)-benzene and 1,4-bis-(β-cyanostyryl)-2,5-dimethoxybenzene. These compounds serve as low molecular weight model compounds for phenylamino-PPV and cyano-PPV,[1] respectively. In keeping with recent publications involving the same materials we refer to the compounds as SP35 and G33, respectively. Table I lists the optical properties of these two compounds in solid poly(styrene) waveguides. Fluorescence half-widths are ~50 nm for both species. Chemical structures are included in Fig. 1. Synthesis of the compounds is described by Hörhold *et al.*[2–4] Optical gain spectra for the compounds in poly(styrene) waveguides are reported by Kretsch *et al.*[5] from amplified spontaneous emission (ASE) measurements. Lasing characteristics of SP35 in toluene solutions, and stimulated emission and excited triplet absorption cross sections for G33 are reported by Henari *et al.*[6,7] The high fluorescence quantum yields. ϕ_f, and large optical gains bode well for use of these compounds in laser devices and or amplifiers.

Kogelnik and Shank[8] first demonstrated the DFB dye laser in a dye doped gelatin film with a photobleached grating. Kogelnik and Shank's theoretical treatment of DFB lasers using coupled wave theory[9] showed that optical feedback is obtained from a pump beam induced spatial modulation of both gain and refractive index.

Figure 1 shows the experimental setup.[10] The pump source is a frequency tripled Nd–YAG laser delivering 33 ps pulses at $\lambda_P = 355$ nm with a 5 Hz repetition rate. A cylindrical lens is used before the beam splitter in order to enable efficient pumping over a narrow stripe on the sample. The spot size of the beam on the film is 2×10^{-3} cm^2 resulting in a maximum pump intensity of 4×10^8 W cm^{-2}.

The pump beam is split by the two prisms P_1 and P_2. The two resulting beams are directed to the film by two corotating mirrors M_1 and M_2. The wavelength of the DFB laser emission will be near the wavelength, λ_L, corresponding to the grating period formed by the pump beams given by[9]

$$\lambda_L = \frac{n\lambda_P}{m \sin\theta} = \frac{2\pi n}{\beta_0}, \tag{1}$$

where λ_P is the pump wavelength, n is the refractive index of the film at the emission wavelength λ_L, θ is the angle of incidence of the pump beams at the sample, and m is the diffraction order. This implies a spatial periodicity of π/β_0 for the grating structure, and the Bragg condition is satisfied for wavelengths with propagation constants close to β_0. Modulation depths for refractive index and gain modulations are expected to be $\Delta n \sim 10^{-4}$ and $\Delta g \sim 10$ cm^{-1}, respectively. For the studies reported here, we use the second order diffraction, $m = 2$, corresponding to angles near $\theta = 35°$ for emission at $\lambda_L = 500$ nm, with $n = 1.6$. The light emitted from the sample is collected by an $f = 50$ mm lens and focused onto an optical fiber bundle (core diameter: 0.6 mm)

TABLE I. Optical properties and lasing characteristics of the model compounds under study in solid poly(styrene) films. Optical gain (ASE) measurements are described in Ref. 5. Solution lasing measurements are described in Ref. 6.

	SP35	G33
Absorption λ_{max} (nm)	390	410
Fluorescence λ_{max} (nm)	470, 490	505
Fluorescence yield, ϕ_f,	0.85	0.65
DFB lasing λ_{max} (nm)	483	525
DFB tuning range (nm)	475–490	510–535
Soln. lasing tuning range (nm)	473–492	⋯
ASE λ_{max} (nm)	487	510
ASE spectral range (nm)	477–501	503–530

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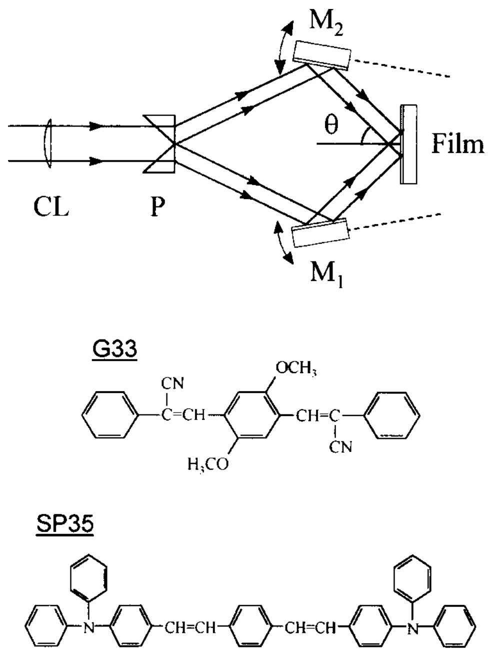

FIG. 1. Schematic diagram of the experimental setup for DFB lasing, forming the grating in the film by means of interference in the pump beams. The chemical structures of the compounds under study are included.

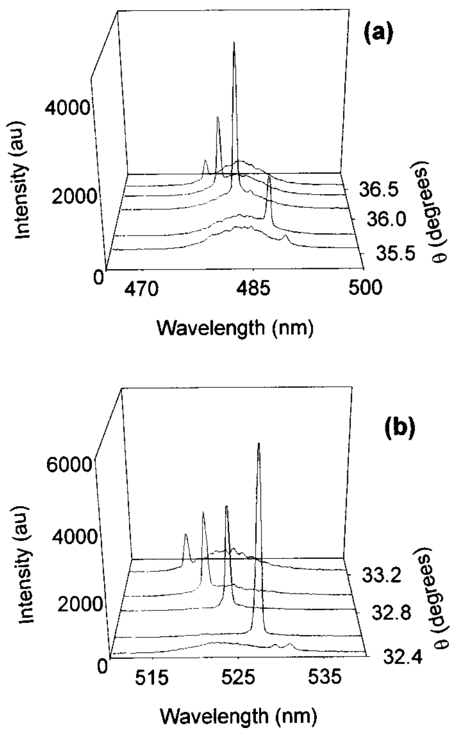

FIG. 2. Laser emission spectra for poly(styrene) waveguides doped with SP35 (a) and G33 (b), demonstrating tuning of the emission wavelength by changing the angle of incidence, θ, of the pump beams on the sample.

coupled to a CCD spectrometer with 0.15 nm resolution. Once laser action is obtained, the emission spectra are monitored as θ is changed.

The samples are produced by spin coating from *ortho*-xylene solution onto silicon wafers with a 1.5 μm thermal oxide overlayer, producing poly(styrene) optical waveguides containing 1.3 wt % of the model compounds. Waveguide characterization was determined using prism coupling[11] experiments. The G33/PS film thickness is 0.69 μm, supporting three guided modes (Ref. 12). TE_0, TM_0, and TE_1. The SP35/PS film thickness is 0.61 μm, supporting TE_0 and TM_0. The SP35 film can support TE_1 but this mode is evanescent into the substrate, and is absorbed by the silicon wafer. Waveguide propagation losses at $\lambda = 632.8$ nm are approximately 2.5 dB cm^{-1}, and no higher than 4 dB cm^{-1} at $\lambda = 514.5$ nm, indicating small ground state reabsorption losses in the fluorescence region.[5]

Figure 2 shows the emission spectra obtained for the doped poly(styrene) waveguides at the maximum incident pump intensity of 4×10^8 W cm^{-2} (1.3×10^{-2} J cm^{-2} at 33 ps pulse duration). We estimate that 30% of the 355 nm pump pulse is absorbed by the samples. Table I includes the characteristics derived from these measurements. Clearly evident are strong laser emission peaks, typically of full width at half maximum (FWHM)=0.6 nm, and a prominent broad spectral background, FWHM=10 nm, attributed to fluorescence narrowing via amplified spontaneous emission.[5]

The samples exhibit a single laser emission maximum, at λ_{max}=483 nm for SP35 doped sample and λ_{max}=525 nm for the G33 doped sample. Laser emission was readily tuned between 475 and 490 nm for SP35 doped films and 510–535 nm for G33 doped films. This broad tuning of the G33 doped film is well evidenced in Fig. 3, showing two simultaneous laser emission peaks with 23 nm separation for a G33 doped waveguide. We attribute these lasing peaks to the two TE modes of the waveguide, TE_0 lasing at 532 nm and TE_1 lasing at 509 nm. Owing to the different propagation constants of the waveguide modes, the modes can only satisfy the Bragg condition at different wavelengths.[13] The effective mode index, $N_{\mathrm{eff},j}$, of the jth order guided mode is related to the propagation constant by $\beta_j = 2\pi N_{\mathrm{eff},j}/\lambda$. The DFB lasing wavelengths [Eq. (1)] are expressed in terms of the effective mode indices using

$$\lambda_{L,j} = \frac{\lambda_P N_{\mathrm{eff},j}}{m \sin\theta} \tag{2}$$

and hence the lower order guided modes (higher N_{eff}) lase at the longer wavelengths.

Based on the expected modulation depths and the Kogelnik and Shanks theoretical treatment,[9] stopbands for these devices are expected to be small ($\sim 10^{10}$ Hz) and the broad ASE background is not suppressed by the grating. All nonlasing guided modes still propagate and can compete for excited states with the lasing mode(s). As the molecular re-

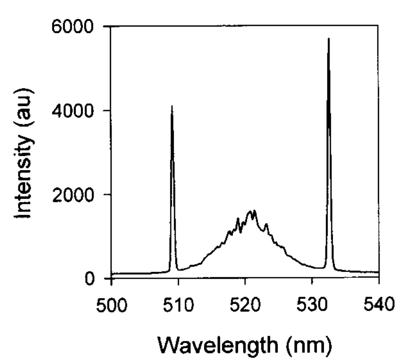

FIG. 3. Emission spectrum of G33 doped poly(styrene) waveguide, displaying two strong laser emission peaks, 532 and 509 nm, attributed to TE_0 and TE_1 guided modes, respectively.

orientation time is much longer than the stimulated emission timescale, only TE polarized emission can occur for transversely pumped films and no TM mode emission is observed. Lasing efficiencies were not determined for these devices.

The SP35/PS waveguide tuning range is in excellent agreement with the toluene solution tuning range reported by Henari *et al.*,[6] and with the positive limits of the optical gain spectum (477–501 nm), determined in Ref. 5 from ASE spectra. The DFB measurements display a small redshift with respect to the ASE measurements. Bearing in mind that the ASE measurement produces higher uncertainties for small optical gains, the DFB and ASE measurements are consistent. This supports the use of the ASE experiment in determining the potential of a material for use as a laser medium. The G33/PS tuning range shows a distinct redshift relative to the ASE measurements in Ref. 5, as indicated in Table I. This redshift is unexplained, as the samples were fabricated from the same batches of materials under identical conditions for both measurements.

Lasing thresholds for the DFB devices are at pump intensities of 7×10^7 and 9×10^7 $\mathrm{W\,cm^{-2}}$ for the SP35 and G33 doped samples, respectively. We note that these threshold intensities are two orders of magnitude higher than intensities required for maximum single pass optical gain in the same materials determined from ASE measurements (Ref. 5). Despite the reduced efficiency of second order diffraction, these large values are yet to be fully explained.

In conclusion, we have demonstrated tunable distributed feedback laser action in poly(styrene) waveguides doped with amino- (SP35) and cyano- (G33) substituted phenylene vinylene model compounds. Comparison of the tuning range with solution lasing measurements and optical gain spectra shows a good agreement for SP35 doped samples.

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