

Subpicosecond pulses from a gain-switched polymer distributed feedback laser

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The emission dynamics of polymer distributed feedback lasers have been studied following excitation by femtosecond laser pulses. The lasers were found to generate pulses as short as 800 fs. The dynamics were investigated as a function of pump power and excitation spot size. Simple rate equations modeling the amplification of spontaneous emission are shown to describe the behavior of the lasing dynamics observed. © 2004 American Institute of Physics. [DOI: 10.1063/1.1767952]

In recent years, the use of conjugated polymers as gain media in lasers and optical amplifiers has been demonstrated. Their large gain cross sections available over a wide spectral range, means that they are promising materials for optoelectronic applications such as broadband amplifiers and tunable lasers.^{1–3} Their ease of processing compared with inorganic materials⁴ means they are a simpler, and potentially inexpensive technology. To date, the main focus of research has been on the reduction of the lasing threshold via improved materials and laser configurations. A reduction in the threshold could help lead to the fabrication of electrically driven organic diode lasers. While amplified spontaneous emission (ASE) and lasing has been demonstrated in a variety of resonator configurations,^{5–8} the temporal dynamics of these optically pumped lasers have not been studied in-depth. The dynamics of these devices could be particularly interesting, since conjugated polymers exhibit large emission bandwidths, leading to their possible use as tunable ultrafast light sources. Recently, picosecond ASE pulses have been observed.^{9,10} In this letter, we report the study of the temporal dynamics and the generation of subpicosecond pulses from a surface-emitting polymer distributed feedback (DFB) laser. We present spectral and temporal characteristics of the emission and rate equations describing the laser dynamics.

The polymer DFB laser consisted of a two-dimensional “egg box” corrugated silica substrate with a grating of period 409 nm in both directions and a peak-trough height of 100 nm. Thin films of poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene vinylene] (MEH-PPV) (thickness ~120 nm) were spin coated onto these substrates. The dynamics of the polymer DFB laser were investigated as a function of both pump intensity and excitation spot size (120, 400, and 600 μm diameter). The experimental setup is shown in Fig. 1. The polymer laser was placed in a vacuum chamber and held under a vacuum of 10^{-5} mbar to prevent contact with water and oxygen. Optical excitation with 100 fs pulses at 510 nm with a repetition rate of 5 kHz was provided by an optical parametric amplifier pumped by a regenerative amplifier. This excitation could be focused by a lens to different spot sizes. An imaging spectrograph with a streak camera (Hamamatsu) detector was used to characterize the emission from the polymer DFB laser. Two lenses

were used to capture, collimate, and focus the emission from the DFB laser into the spectrograph. The temporal response of the streak camera to the 100 fs excitation pulses was measured to determine the time resolution of the instrument. All pulse lengths are quoted as the FWHM of the pulse dynamics. Laser linewidths are limited by the resolution of the spectrograph.

Figure 2 shows the spectral and temporal evolution of the emission from the polymer DFB laser for spot sizes of 400 and 600 μm diameter ($1/e^2$ measurements), to illustrate the typical behavior observed. Figure 2(a) illustrates the emission spectrum for the 400 μm excitation spot. For low excitation densities the emission is centered at 620 nm, with a linewidth of ~10 nm, and is attributed to Bragg scattered ASE.¹¹ At an excitation density of ~2.8 $\mu\text{J cm}^{-2}$ (which corresponds to an exciton density of order 10^{18} cm^{-3}) a feature appears on the spontaneous emission background. This is the lasing threshold and as the excitation intensity is increased above this level the emission narrows to a laser peak with a central wavelength of 620 nm and a linewidth of 1.5 nm (FWHM).

In Fig. 2(b) the corresponding time dynamics of these pulses are shown, with measurements taken using the shortest possible time window of 20 ps on the streak camera. A longer-lived fluorescence lifetime for MEH-PPV of 180 ps was observed with a longer time window but is not shown here, this is consistent with other measurements of films of MEH-PPV.¹² Below threshold (2.1 $\mu\text{J cm}^{-2}$), the pulse length (FWHM) is broad with a fast decay component of 2.5 ps and a slow decay component of 8 ps. As the excitation intensity is increased above the lasing threshold the slower

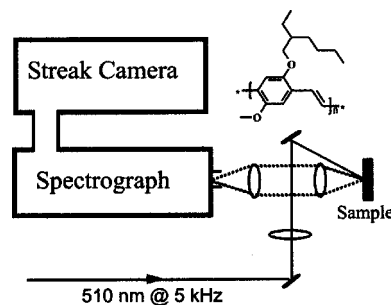


FIG. 1. Experimental setup for investigating the DFB laser emission and the chemical structure of MEH-PPV.

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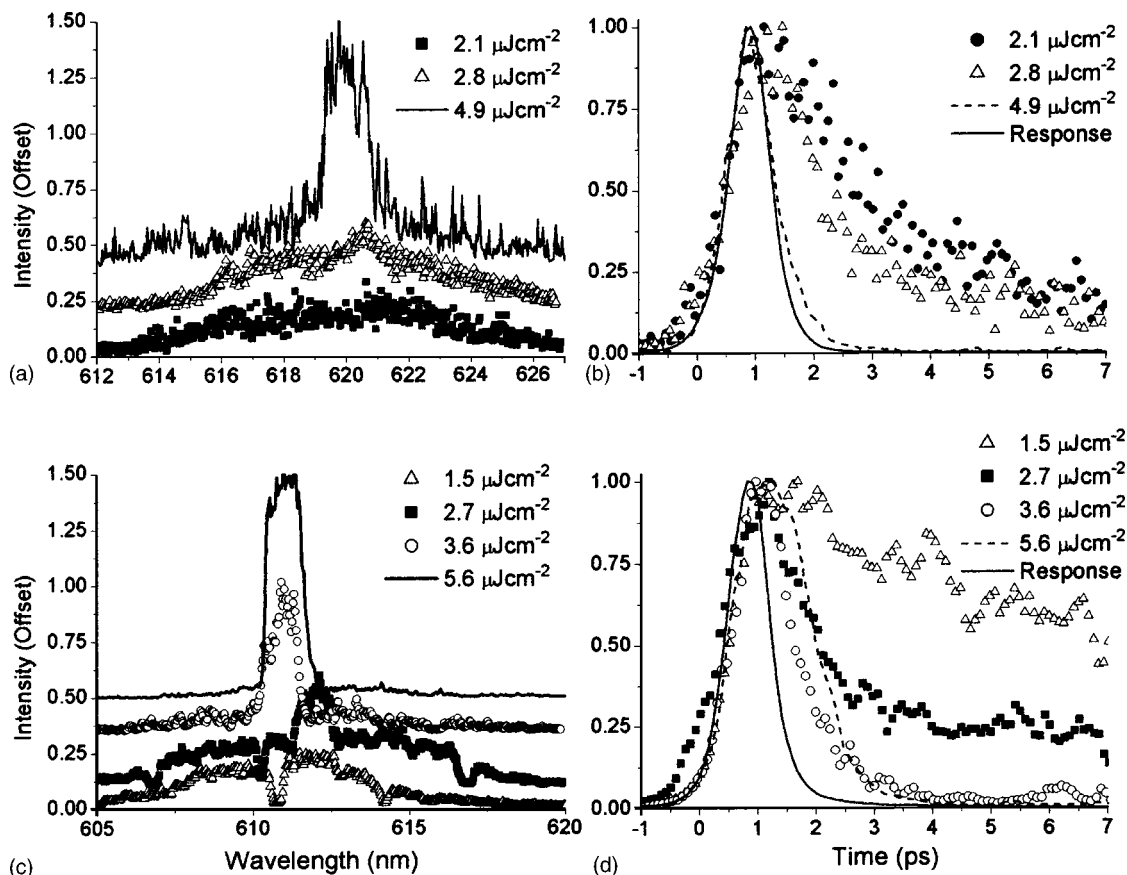


FIG. 2. Spectral and temporal dynamics for excitation spot sizes of 400 μm (a), (b) and 600 μm (c),(d). Spectra in (a) and (c) are offset for clarity, pulse dynamics are normalized.

decay component disappears and the pulse length shortens towards the response of the streak camera (750 fs, FWHM), with a shortest pulse of 850 fs observed for an excitation density of 4.9 $\mu\text{J cm}^{-2}$.

Figures 2(c) and 2(d), illustrate the spectral and temporal kinetics for the 600 μm excitation spot. In Fig. 2(c) the Bragg scattered ASE background is seen at low excitation densities. A laser peak emerges from this at a pump intensity of 2.7 $\mu\text{J cm}^{-2}$. Increasing the excitation density further above this threshold, narrows the laser emission to a peak at a wavelength of 611 nm with a linewidth of 1.5 nm (FWHM). The differences in wavelength of the emission in Figs. 2(a) and 2(c) are due to slightly different film thicknesses used for the two sets of measurements. Figure 2(d) shows the time evolution of the laser pulses. At a low excitation density (1.5 $\mu\text{J cm}^{-2}$) there is a fast component of 2 ps and a slow component of 15 ps. The slow component disappears as the excitation density is increased above the threshold of 2.7 $\mu\text{J cm}^{-2}$ and the shortest pulses observed are at 3.6 $\mu\text{J cm}^{-2}$, with a pulse length of 1.4 ps. After the pulselength narrows towards the response of the detector, higher excitation densities produce broader pulses (2 ps for 5.6 $\mu\text{J cm}^{-2}$). This broadening of pulses at higher excitation densities was observed for all spot sizes.

Figure 3 shows the shortest pulses observed for the different excitation spot sizes. The shortest pulses observed were 800 fs (120 μm spot) and 850 fs (400 μm spot), each observed at about twice the intensity of the laser threshold. These shortest pulses are close in duration to the response time of the system and deconvolution of the signals shows that the pulse length is shorter than the instrument response

of 750 fs. The shortest pulse from the 600 μm excitation spot was ~ 1.4 ps.

Our results show a clear lasing threshold, above which both the spectral and temporal kinetics change. This threshold was found to be $\sim 2.7 \mu\text{J cm}^{-2}$ and independent of pump spot size. Below threshold, the ASE dynamics are faster than the spontaneous emission lifetime of ~ 180 ps. Taking 800 fs as an upper limit on the pulselength gives an upper limit on the time-bandwidth product of $\Delta\tau\Delta\nu=0.7$, or ~ 1.5 times transform limited. We note that the 600 μm spot size produced longer pulses than the two other smaller excitation spots. This effect may be related to the fact that the 600 μm excitation region is much larger than the estimated scattering length of the feedback ($\sim 200\text{--}300 \mu\text{m}$), which could result in reduced temporal coherence of the emitted light.

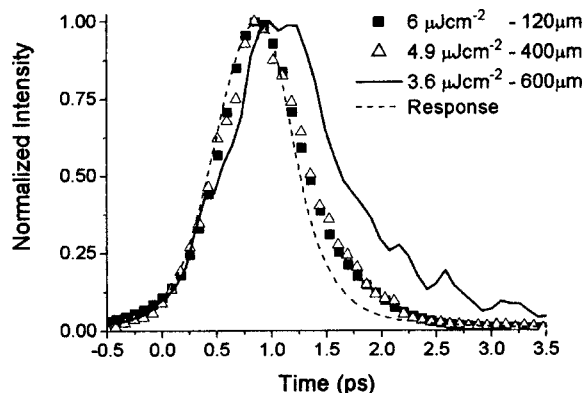


FIG. 3. Shortest pulses from different excitation spot sizes.

In order to model the lasing dynamics of the gain-switched DFB laser we used simple rate equations (similar to those described in Ref. 13) for the upper state population density $N(t)$ and photon density $S(t)$ within the laser.

$$\frac{dN}{dt} = -S(t)V_{gr}[\sigma_{se}N(t) - \alpha_{abs}] - \frac{N(t)}{\tau_{exc}}, \quad (1)$$

$$\frac{dS}{dt} = S(t)\Gamma V_{gr}[\sigma_{se}N(t) - \alpha_{abs}] - \frac{S(t)}{\tau_{ph}}. \quad (2)$$

The dynamics of the upper state population $N(t)$ [Eq. (1)] includes only terms describing the stimulated emission rate and the spontaneous exciton decay. The 100 fs excitation pulse is assumed to produce an instantaneous population inversion (gain-switched operation). V_{gr} is the group velocity of light in the waveguide, taken to be $\sim 2 \times 10^{10}$ cm s⁻¹, α_{abs} the absorption loss measured as 20 cm⁻¹ and τ_{exc} is the exciton decay time constant of 180 ps. The initial population density $[N(0)]$ values were calculated from the energy of the excitation pulses and the initial photon density $S(0)$ was taken to be 0.001% of this value, to approximately represent the number of photons spontaneously emitted into the laser mode within the duration of the pump pulse. The pulse buildup time varied slightly for different values of $S(0)$ but the calculated pulse widths varied by less than 5% over a change of six orders of magnitude for the initial photon density.

The dynamics of the photon density [Eq. (2)] includes terms describing the stimulated emission and the lifetime of the photons in the resonator. The photon lifetime τ_{ph} , can be expressed as a function of the group velocity and a loss factor combining absorption and scattering of photons, $\tau_{ph} = 1/(V_{gr} \cdot \alpha_{loss})$, where α_{loss} represents the losses from the resonator including both absorption and scattering of the photons. The light confinement factor Γ of the 2D-DFB laser (applied to photon density) was calculated to be 0.4.¹⁴ In these equations the fitting parameters were the stimulated emission cross-section σ_{se} of MEH-PPV in the DFB laser and the α_{loss} term. The loss term was taken to be 50 cm⁻¹, more than twice the absorption loss because Bragg scattering is likely to be a larger loss contribution in the DFB grating. A value of 3×10^{-16} cm⁻² for σ_{se} was found to give the best agreement with the experimental data. This value is comparable to 8×10^{-17} cm⁻² from Ref. 15.

Following the instantaneous excitation, the emission evolves from this initial population of the emitting state. The model, as shown in Fig. 4, successfully explains the pulse narrowing but does not, however, account for the rebroadening seen at higher intensities. This latter effect may be related to the onset of independent single grating lasing,^{8,9} which has both lower spectral and spatial coherence than the laser mode supported by the two-dimensional structure.

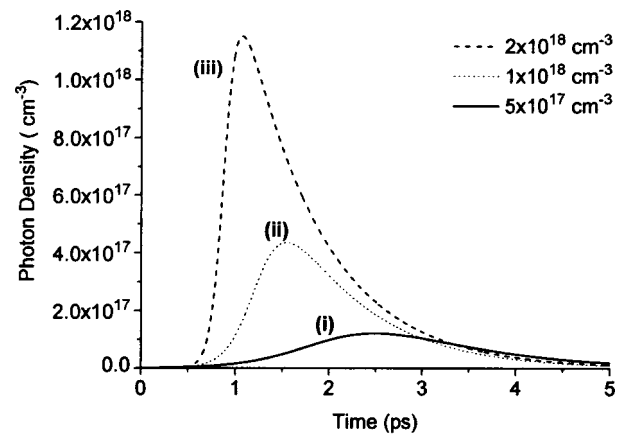


FIG. 4. Theoretical plots of photon density for initial population inversions $N(0)$ of (i) 5×10^{17} cm⁻³, (ii) 1×10^{18} cm⁻³, and (iii) 2×10^{18} cm⁻³.

In conclusion, we have demonstrated that subpicosecond pulses can be generated by a gain-switched MEH-PPV distributed feedback laser. These results can be explained by the growth of amplified spontaneous emission as shown by simple rate equations.

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