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Picosecond gain switching of an organic semiconductor optical amplifier

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All-optical switching of an individual pulse within a sequence of amplified pulses is demonstrated in a conjugated polymer optical amplifier. The switching was achieved using a control pulse resonant with the excited state absorption. An extinction ratio of ~5.5 dB was observed, while the intensities of the remaining pulses in the sequence, spaced at 50 ps intervals, were unaffected. A pump-probe study was performed and showed full gain recovery within 2 ps. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2883975]

A great deal of interest has been shown in conjugated polymers in recent years as gain media for optoelectronic devices. They have high gain over a broad bandwidth,[1–3] are simple to process leading to low manufacturing costs, and has the potential to be electrically pumped.[4] Optoelectronic devices made from conjugated polymers would be readily compatible with polymer integrated circuits and polymer optical fibers which are favored for short haul data transmission networks.[5,6] Conjugated polymers have been demonstrated as optically pumped, high gain laser devices,[7–10] as well as amplifier devices in solution[11,12] and in the solid state.[13–15]

In addition to lasers and amplifiers, all-optical switching devices are essential building blocks for future ultrahigh speed optical communications networks, integrated circuits, and signal processing. There has been a long standing quest for all-optical switching in organic optoelectronic devices. The main approach investigated has been to use the nonlinear refractive index associated with $\chi^{(3)}$ nonlinearities.[16–20] However, in spite of the large nonlinearities of these materials, the losses associated with one or two photon absorption mean that satisfying the relevant figures of merit required in devices has proved challenging.[17,19,20] We have therefore pursued an alternative approach and investigated using optical control of gain as a possible route to all-optical switching. It has been shown in pump-probe experiments that transmission changes of a few percent can be achieved by a control light pulse.[21,22] In one case, the gain was "dumped" so it was not available subsequently[21] and in the other case gain control was limited to selected regions of a phase separated polymer blend.[22] It has also been shown that a control pulse can change the output of a conjugated polymer laser.[23,24] The switching of a laser can take advantage of the fact that lasers have a threshold so that a small change in gain can lead to a very large change in the output. Amplifiers do not have a threshold in the way that lasers do and so optical switching of an amplifier is a more challenging problem. In this letter, we show high speed optical switching of a polymer optical amplifier, use it to turn off the amplification for one pulse in a pulse train, and measure the gain recovery time.

The switching is achieved by gain modulation in a conjugated polymer to implement all-optical switching. The switching concept is shown in Fig. 1. Figure 1(a) shows the gain obtained by stimulated emission from the lowest excited state S_1 and the amplified output pulse. The control pulse is matched to the absorption in the S_1 state of the material and when it is applied, the excitons in the S_1 state are excited to a higher state S_n, as shown in Fig. 1(b). This reduces the population of the S_1 state and reduces gain so that the output is just the low powered input pulse. In this manner, the amplification of a pulse can be switched off. Relaxation from the S_n state to S_1 then leads to gain recovery.

The amplifier structure is shown in Fig. 2(a). Gratings were etched into the silica substrate by reactive ion etching, to couple the signal into and out of the device. The gain medium was a conjugated copolymer consisting of fluorene and benzothiadiazole units and its absorption and photoluminescence spectra are shown in Fig. 2(b). It was deposited by spin coating from toluene solution to make a film of 600 nm thick on top of the grating structure. The active region of the amplifier lies between the grating couplers and was pumped at a wavelength of 497 nm, which had an absorbance of 0.2, at a repetition rate of 5 kHz. The signal pulses were at 580 nm and, like the pump pulses, were generated by an optical parametric amplifier which was pumped by an amplified Ti:sapphire laser system at 5 kHz. The pump, signal, and control pulses were stretched in a TF-10 glass block from 100 fs to 10 ps to match the resolution of the streak camera, which was used to record the output pulses. A sequence of

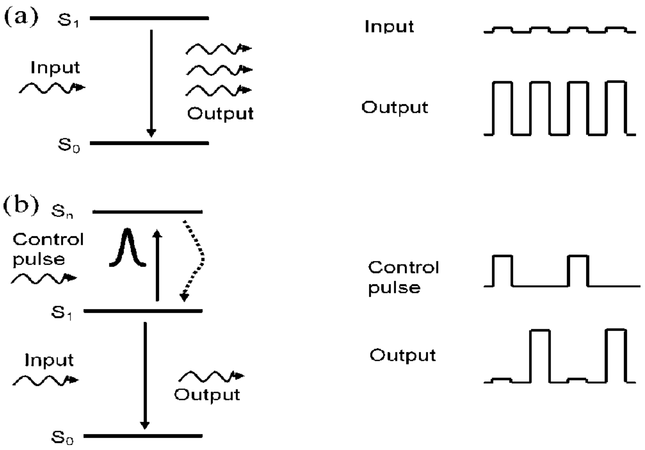

FIG. 1. Principle of the on-off switching presented in the form of an energy level diagram and pulse sequence. (a) Without control pulse: input pulses are amplified by stimulated emission. (b) With control pulse: the control pulse reduces population in the S_1 excited state so that no amplification occurs and the input pulse passes unchanged.

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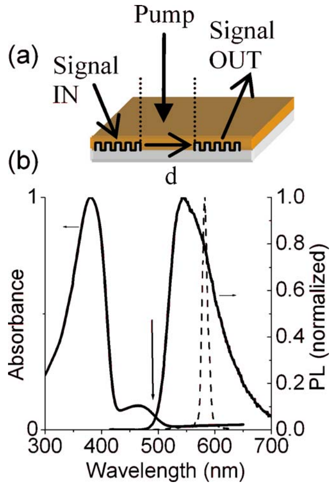

FIG. 2. (Color online) (a) The amplifier structure. The signal couples into and out of the polymer waveguide via the gratings and the region in between is optically pumped. (b) The absorption and luminescence spectra of the copolymer film. An arrow indicates the spectral position of the pump at 497 nm, the dashed line is the probe signal.

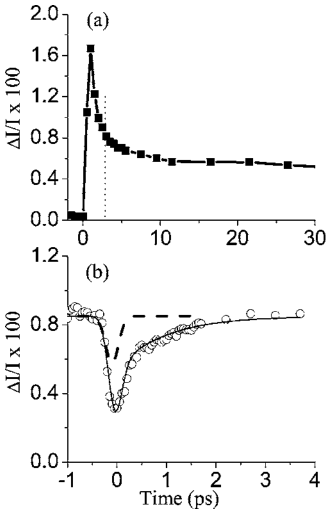

FIG. 4. (a) Increase of the probe signal against the pump-probe time delay without a control pulse present. The dotted line indicates where the delay stage for the pump pulse was paused and the effect of the control pulse was studied. (b) Change of the probe signal vs the control-probe time delay (symbols), the dashed line is the instrument response function and the solid line is a biexponential fit to the recovery kinetics (see text).

signal pulses was obtained with a partial reflector and a mirror. The polarization of the signal was rotated by a waveplate so that it can be deflected off the input path by a Glan polarizer and directed to the sample. A control pulse at 800 nm was applied to the pumped region.

Figure 3(a) shows the unamplified signal pulses (open symbols) and solid symbols show the same pulse sequence amplified with a pump. The middle pulse was amplified by a factor of 4, and the remaining pulses by a factor of 2. A control pulse with an energy of 1 μJ was applied at 800 nm to the middle pulse. When the control pulse is applied, the amplified middle pulse drops in intensity by a factor of 3 and is only 30% higher than the nonamplified pulse. When the control pulse is turned off, the amplification of the middle pulse returns to its original level, as shown in Fig. 3(b). This is vital as the output level should not be affected after the control pulse has been removed. Moreover, the control pulse has no effect on the two neighboring pulses.

The gain recovery time after the switching was measured using a transient absorption technique. The polymer amplifier was pumped with 100 fs pulses at 400 nm and probed with delayed 50 fs pulses at 550 nm. The pump pulses were synchronously chopped at 2.5 kHz and the increase in probe intensity due to stimulated emission from the S_1 state was measured as a function of the time delay between the pump and probe pulses using a lock-in amplifier. This fractional increase in probe intensity $\Delta I/I$ is shown in Fig. 4(a). Decay of the probe amplification factor within the first 5 ps is observed, which is due to the buildup of amplified spontaneous emission. With the probe pulse maintained at 3 ps after the pump pulse, the effect of the 100 fs control pulse at 800 nm on the $\Delta I/I$ signal as a function of the control-probe time delay was investigated. The control pulses cause a decrease of the $\Delta I/I$ signal, as observed in Fig. 4(b) because a substantial part of the S_1 population is pushed to the S_n state. Amplification recovers as the population relaxes to the S_1 state.

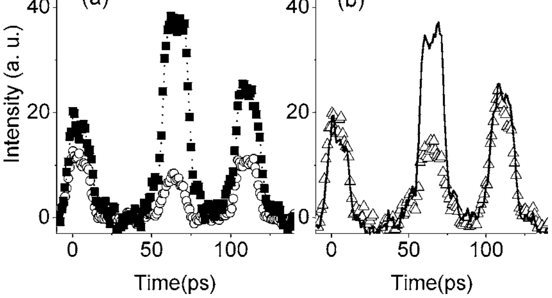

FIG. 3. (a) Unamplified (open symbols) and amplified pulse sequence (solid symbols) detected at the output of the 400 μm waveguide. Energies of the pump and the signal pulses were 221 and 0.13 nJ. (b) Amplified pulse sequence when control pulse with an energy of 1 μJ matched in time with the middle pulse (open symbols) and the amplified pulse sequence after the control pulse was turned off (solid line).

An important feature of the result is that there is full and rapid recovery of the gain in 2 ps. This is in contrast to a previous report of polyfluorene dispersed in a polymethylmethacrylate matrix in which 80% of the gain recovered in 3 ps.[22] The incomplete recovery followed a $t^{-1/2}$ power law and was attributed to geminate recombination of intrachain charge pairs. In contrast, the recovery in Fig. 4(b) can be fitted to a biexponential decay function with time constants of $\tau_1=(50\pm20)$ fs and $\tau_2=(1\pm0.1)$ ps but not to $t^{-1/2}$. The preexponential factors for the two components were 0.88 and 0.12, respectively. The fast component can be assigned to internal conversion from the S_n to the S_1 state. In this process, the electronic $S_n \leftarrow S_1$ excitation energy is converted into vibrational energy, which subsequently is distributed over all intramolecular vibrational modes. Some excess energy stored in optically active vibrational modes is dissipated on a longer time scale by heat transfer to unexcited molecules and can explain a 1 ps component in the gain recovery. Our results show that the use of the copolymer helps to reduce charge separation and to achieve full gain recovery. It takes 2 ps for the gain to recover fully, therefore, the highest possible switching rate for this copolymer is 500 GHz.

In summary, we have demonstrated all-optical switching of an optical amplifier. We have shown that the effect can be applied to one pulse in a sequence of amplified pulses without affecting the amplification of the remaining pulses. The switching effect had a fast and full recovery in 2 ps, which is comparable to fast recovery times observed in inorganic semiconducting optical amplifiers.[25–27] Our results demonstrate an alternative to nonlinear optical switching and show that optical switching and amplification functions can be combined in a single, compact, and easily fabricated device.

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