

Ultrafast and Nano Optics

(超快和纳米光学)

Xinping Zhang and Yanrong Song *et al.*



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Ultrafast and Nano Optics

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Introduction

This is the proceedings of the International Symposium on Ultrafast and Nano-Optics 2007 (UNO2007). This symposium was held on 28–31 October 2007 in Beijing University of Technology. More than 45 scientists came to the symposium from 7 countries and gave presentations, reporting their recent progresses in the research fields of ultrafast and nano-optics, as well as in other related topics. This proceedings includes 32 full papers that have been presented at this symposium.

The symposium was very successful in stimulating extensive discussions in the focused topics in optics. We acknowledge all of the scientists that have attended this symposium for their contributions and hope that this meeting will be helpful for establishing close communications and collaborations between these active research groups.

Xinping Zhang
Yanrong Song
April 2008

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Section I

Ultrafast Optics and Applications

Polymer Optical Fiber Amplifiers Based on Conjugated Fluorene Oligomers

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Abstract: We demonstrate polymer optical fiber (POF) amplifiers operating between 440 nm and 480 nm using POFs doped with a series of fluorene oligomers. The optical and morphological characteristics of doped PMMA films and POFs were studied and demonstrate that the oligomers were well dispersed within the PMMA matrix and survive the PMMA polymerization process to produce POF amplifiers with net gain on the order of 0.07 dB.

Introduction

Polymer Optical Fibers (POFs) have attracted much attention recently owing to their unique properties, such as flexibility, low production cost and ease of optical coupling^[1,2]. Their transmission and dispersion properties do not rival that of silica fibers, as standard poly(methylmethacrylate) (PMMA) POF attenuation is on the order of 100 dB/km, compared with <1 dB/km for silica fibers. However, their ease of use and low-cost nature pave the way for their application in short-haul data-communication networks where many data-links are required and where speed of transmission and attenuation are not critical.

The possibility to dope POFs with functional materials opens a wide range of new applications such as all-optical amplifiers, switches, sensors, scintillators and so on. In this work, we demonstrated POF amplifiers (POFAs) which operated in the blue spectral region (440–480 nm). These were produced by doping the PMMA POF with a series of conjugated organic oligomers, similar to the commonly-studied poly(9,9 dioctylfluorene) (PFO) (Fig.1). The motivation behind doping the POFs with fluorene oligomers is manifold. Fluorene polymers are known to be stable gain materials, with high gain coefficients and low thresholds^[3,4]. It is likely that the oligomers will have similar properties. Furthermore, recent studies have shown that isolated chains of PFO in PMMA can support short-lived intrachain charges^[5]. These intrachain charged states can be used to switch the gain in polyfluorene materials as their absorption overlaps spectrally with the

stimulated emission^[6]. Therefore, attainment of isolated conjugated molecules in POFs could lead to all-optical gain media capable of ultrafast switching. However, conjugated polymers such as PFO do not mix well with aliphatic polymers such as PMMA^[7]. This immiscibility would lead to aggregation of the PFO in the optical fibres, reducing the switching applicability and leading to large losses in transmission due to scattering^[1,2]. Conjugated oligomers, on the other hand, are likely to be more miscible in PMMA producing homogeneous films and POFs with isolated conjugated molecules in the PMMA matrix.

Experimental

PMMA was obtained from Sigma Aldrich and used without further purification for the thin-film studies. POFs were made at LUCEAT s.p.a. following standard procedures. The fluorene oligomers, tri-(9,9-dioctylfluorene) (here referred to as trimer), penta-(9,9-dioctylfluorene) (pentamer) and hepta-(9,9-dihexylfluorene) (heptamer), and polymer PFO were obtained from American Dye Source, Inc and were used without further purification. The structures are shown in the inset in Fig. 1.

For optical measurements on the pure materials, 100 nm to 130 nm thick films were made by spin-coating 50 mg/mL toluene solutions onto polished synthetic quartz (Spectrosil B) substrates. Blend films of oligofluorene: PMMA in 10% or 16% by weight ratios were made by spin-coating toluene solution (50 mg/mL of PMMA) onto quartz or glass substrates, producing films ~200 nm thick. The film thicknesses were measured with a Dektak Surface Profilometer.

POFs were produced using an adapted preform-drawing technique at LUCEAT s.p.a.^[8]. The dopant oligofluorenes were added to liquid methylmethacrylate monomer (MMA) at a concentration of 0.02% by weight to produce preforms (~4 cm in diameter) which were then drawn into fibers using a standard technique^[2]. The resultant doped POFs have diameters of 1 mm and are clad with a fluorinated polymer. The ends of the POFs were polished prior to measuring using polishing paper.

Steady-state photoluminescence (PL) spectra were measured using excitation from either an argon-ion laser at 3.42 eV (362.8 nm) (films) or a diode laser at 3.31 eV (375 nm) (doped POFs). The emitted light was collected using a fiber bundle and dispersed into a spectrograph with a charge-coupled device (CCD) detector. Absorption measurements on films were performed using a Unicam UV 500 UV-Vis spectrometer.

Attenuation measurements of the POFs were performed using the standard cut-back method^[2] on fibers 5 m long. Fibers longer than 5 m are usually measured using this method, however attenuation of the doped fibers is much higher than that of the pure PMMA fibers and so shorter sections were used to avoid saturation of the measurement. Attenuation of the fibers was also measured using side-illumination fluorescence spectroscopy (SIF), which has been previously been used to measure the attenuation of POFs doped with PSQ and HSQ dyes^[9]. In this second method, several PL spectra of the doped-POFs are recorded. To start with, the PL spectra are measured by exciting the POF normal to the POF axis with detection also at normal incidence, producing a spectrum exhibiting minimal self-absorption. Other measurements were also performed with

the excitation still normal to the POF axis but with the detection parallel to the axis. Detecting light from the end of the POF but exciting at various distances from the end of the POF permits a quantitative measurement of the self-absorption to be made. If we assume that the PL produced by the excitation is a plane-wave source, we can take two PL spectra measured at different distances from detection and, using the Beer-Lambert law in Eq. (1), we can estimate the attenuation coefficient spectrum.

$$I_1(\lambda, z_1) = I_0(\lambda) \exp(-\alpha z_1) \quad (1)$$

Converting the units of the attenuation coefficient, α from cm^{-1} into dB/km ($A = 10^6 \alpha \lg[e]$), we can compare the results from the two measurements. The SIF measurement is indirect and is performed on short segments of fiber. In these short segments, the mode distribution of the waveguide has not yet reached equilibrium (this can take tens of meters in PMMA POFs^[2]), the attenuation under these circumstances is higher than at equilibrium mode distribution (measured using the cut-back method). Therefore, the SIF method will produce an upper limit of attenuation in the POFs. In a real POF network the transmitted light will be at equilibrium mode distribution and so the cut-back method is more applicable. However, most of the measurements of POFs in this study are performed on short segments where the SIF method is more realistic. Here, we present results obtained using both methods, however we use the SIF results when calculating the overall gain values of the POFs.

We have also performed conventional pump-probe transient absorption spectroscopy^[10] on both films and doped POFs. In this experiment, pump pulses at 3.18 eV (390 nm), the second harmonic of the output of a CLARK Ti:Sapphire regenerative amplifier with a repetition rate of 1 kHz and a pulse length of 150 fs, are focused to a 230 μm diameter spot, giving an excitation energy density between 0.1 mJ/cm^2 and 1 mJ/cm^2 . The probe beam is a white light continuum 1.22–3.10 eV (400–1100 nm), generated by focussing the CLARK amplifier output onto a sapphire plate. The pump and probe beams are spatially overlapped on the sample (for the POFs they are overlapped at the front end of the POF). The probe beam intensity used in the experiment is kept deliberately low.

Results

Fig. 1 shows the absorption and emission spectra of the oligomers and polymer thin films compared with the same materials when doped into a PMMA matrix at a concentration of 16% by weight. Increasing the oligomer length causes a red-shift of the spectra, consistent with a reduction of the effective optical band-gap and the particle-in-a-box model^[11, 12]. Upon doping of the active materials into PMMA (dashed lines), it can be seen that the absorption spectra do not change significantly. However, we do observe slight changes in the relative intensity of the vibronic bands of the PL spectra, especially in the trimer film. These changes could be due to self-absorption effects or could be due to changes in the molecular-morphology of the oligomers when doped in to the PMMA matrix^[13]. As no large changes are observed in either the PL or the absorption spectra, we consider the primary neutral excitation in the oligomers to be similar in both the pristine and the blend films.

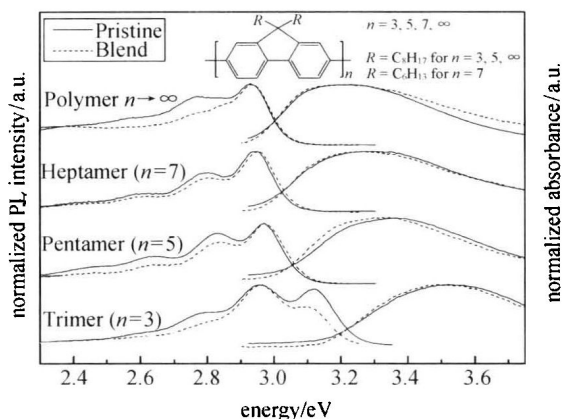


Fig. 1 Normalized PL (left) and absorbance (right) spectra of pristine (solid lines) and oligomer/polymer:PMMA blend (dashed lines) films made up of a fluorene trimer, pentamer, heptamer and polymer (ascending order). The spectra have been vertically shifted for easier comparison. Inset shows chemical structure

To characterize the degree of dispersion of the oligomers in PMMA, the surface topography of the thin-film blends was measured using scanning force microscopy (SFM), as shown in Fig. 2. The height scales of images Fig. 2(a) to Fig. 2(c) are the same and thus we observe an increasing degree of phase separation as the mass of the oligomer increases. For comparison, Fig. 2(d) shows an SFM image of a blend film made from PMMA:PFO (16%wt). This film shows considerable phase-separation, as has previously been reported^[7]. The trimer:PMMA blend film [Fig. 2(a)] has a root mean square surface roughness of only 0.4 nm and shows no evidence of phase-separation on

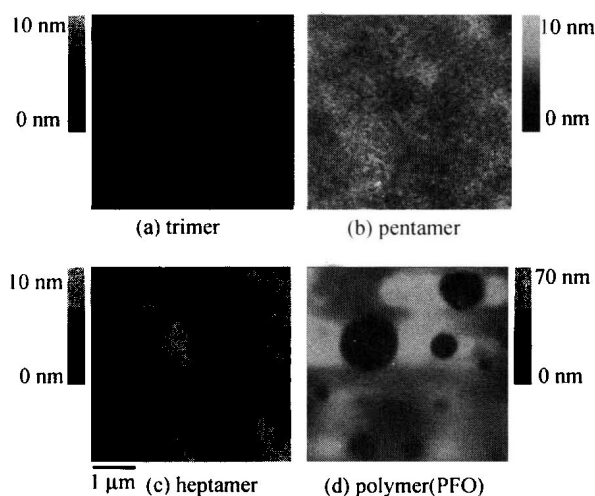


Fig. 2 Tapping mode scanning force microscopy (SFM) images of the films spin-cast from 16%wt blends of the four fluorene materials with PMMA (a) trimer:PMMA, (b) pentamer:PMMA, (c) heptamer:PMMA, (d) polymer:PMMA. The length and height scales are marked on the figure

length scales of 5 μm . The heptamer:PMMA [Fig. 2(d)] blend film however shows evidence of phase-separation, with inclusions (presumed to be heptamer-rich) detected having an average diameter of ~ 200 nm.

Polymer optical fibers were doped with various oligofluorenes by dissolving the oligofluorene chromophores in liquid MMA before the polymerisation process and then fabricating the preform and POF. The concentration of dopant material in the POF was set at a deliberately low level (0.02% by weight) to ensure that aggregation of active material did not occur during the POF fabrication procedure and to ensure solubility of the dopant in liquid MMA. An image of the doped POFs is shown in Fig. 3. Note the photoluminescence exiting the fibres is excited via ambient room light that is incident through the walls of the fiber.

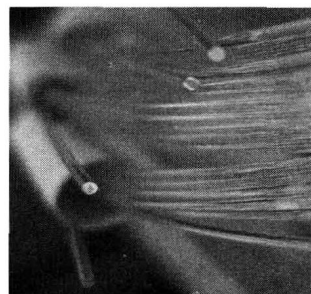


Fig. 3 Photograph of POF doped with (in ascending order) fluorene trimer, pentamer, heptamer and polymer

The PL and attenuation spectra of the oligofluorene-doped POFs are shown in Fig. 4. Part (a) of the figure shows attenuation measured using two different techniques. The lower set of curves (with energies between 1.80 eV and 2.75 eV) were measured using a standard cut-back method. We note that for energies above 2.07 eV, the heptamer-doped

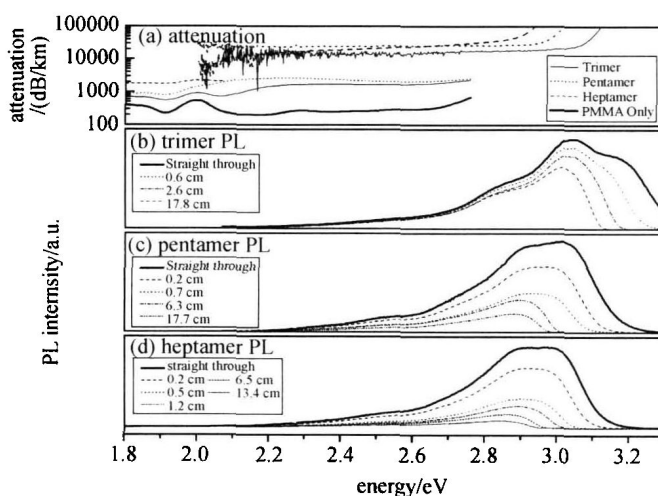


Fig. 4 (a) Attenuation spectra of POFs doped with trimer (thin solid line), pentamer (dotted line) and heptamer (dashed line). PMMA-only POF attenuation shown for comparison (thick solid line). Two measurements were performed and are compared. Those plotted over 1.80–2.75 eV were measured using the cut-back method. Here the heptamer-doped POF was only measured to 2.07 eV due to saturation at higher energies. Curves plotted between 2.00–3.20 eV are measured using side-illumination fluorescence spectroscopy. Parts (b) to (d) show PL spectra for various distances between excitation and detection (as described in the text) for (b) trimer-doped POF, (c) pentamer-doped POF and (d) heptamer-doped POF

fiber attenuation could not be measured due to saturation of the measurement. The attenuation spectrum of a standard (undoped) PMMA POF is shown for comparison. Using this detection measurement the attenuation is well below 5000 dB/km for all samples and there is a systematic increase in attenuation with increasing oligomer length.

The upper set of curves (recorded over an energy range of 2.00 eV to 3.20 eV) were measured using side-illumination fluorescence spectroscopy (SIF). The resulting PL spectra are shown in parts (b) to (d) of the Fig. 4. The SIF method results in higher attenuation values, which reflects the fact that short segments of POF do not reach an equilibrium mode distribution.

To determine the gain properties of the doped optical fibres, femto-second transient absorption spectra were measured. Fig. 5 shows the spectrum of the different doped POFs (filled circles, left axis) along with blend films (open circles, right axis). The spectra are similar to those measured elsewhere for similar materials^[14, 15] and so we assign the positive signal (2.5–2.8 eV) to stimulated emission (note that it also overlaps with the PL spectra in Fig. 1) and the negative signal in the near infrared to absorption from the first excited singlet state (S_1) to a higher-lying singlet state (S_n). The spectra for the POFs and the blend films are similar.

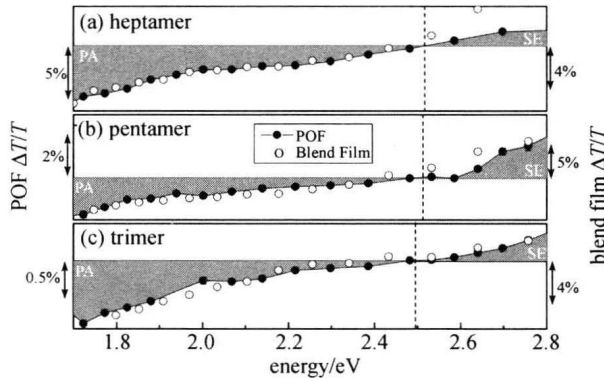


Fig. 5 Differential transient absorption spectra at 2ps after excitation. Open markers denote blend film spectra (right-hand scale), filled markers denote doped POF spectra (left-hand scale), for (a) the heptamer, (b) the pentamer and (c) the trimer

To estimate the gain of the samples, we use the following expression for the enhancement of the signal (or probe light) due to the pump

$$E = 10 \lg \left(\frac{\Delta T}{T} + 1 \right) \quad (2)$$

where E is the enhancement coefficient (in dB), d is the length of the POF (in km) and $\Delta T/T$ is the differential transmission as shown in Fig. 5. Using a value of the attenuation coefficient measured using the SIF method from Fig. 4, we can estimate the gain coefficient in the fiber (G) using $G = E - A$, where A is the attenuation coefficient. This results in a maximum gain of 0.065 dB for a 2 mm POF doped with the oligofluorene pentamer at

2.9 eV. We note that the gain increases with fiber length for fibers up to 2 cm, after which the gain begins to reduce (data not presented). Furthermore, the enhancement (and therefore gain) scales with pulse energy^[16]. Finally, we note that these gain values are a lower limit for the oligofluorene doped POFs as we have used the attenuation values provided by the SIF measurement. In an ideal POF network, the radiation is at equilibrium mode distribution and therefore the attenuation will be lower and the gain higher.

Discussion and conclusions

We have demonstrated that POFs doped with conjugated oligomers can act as optical amplifiers, which may find important applications in short haul POF networks. The motivation for doping the POFs with conjugated oligomers results from the high-gain properties and optical switching capabilities of isolated chains of the conjugated fluorene polymer, PFO^[3–6]. Films of PMMA blended with PFO undergo strong phase separation driven by the large enthalpy costs of mixing^[7] and so we have chosen to explore doping fluorene oligomers into PMMA, as such systems are anticipated to form significantly more homogeneous films without attendant problems of phase-separation or aggregation. Such aggregation or phase separation would almost certainly result in strong optical scattering and would also suppress the fast switching phenomena previously observed when PFO molecules were dispersed in PMMA at low concentrations.

Blending the oligomers with PMMA showed that at concentrations as high as 16% by weight there was little phase-separation in the trimer and pentamer blends, indicating that the small chain-length oligomers are well mixed with the PMMA. The POFs doped with 0.02 wt% active oligomer had attenuation well below 5000 dB/km, again indicating a low degree of aggregation between the oligofluorene molecules. The transient absorption measurements of doped-POFs were similar to the blend films. These findings suggest that the active oligomers survive the harsh PMMA polymerisation process.

Unfortunately, the values of gain reported here are relatively low. These values are lower than those previously reported in POF amplifiers doped with Rhodamine B (27 dB for 50 cm^[17] or 23 dB for 69 cm^[18]), Rhodamine 6G (18 dB for 8 cm)^[19] or EU(DBM)₃ Phen (5.7 dB for 30 cm^[20]). However, the values reported in the literature were measured with high pump energies, on the order of 300 μ J–95 J per pulse (using 5–10 ns pulses)^[17–20]. We measured gain with pump energies of only 400 nJ per pulse (using 150 fs pulses). As enhancement increases linearly with pump energy, we expect to observe much higher gain values at the large pulse energies used in the literature. Unfortunately, we cannot verify this with the equipment available.

In conclusion, we have demonstrated that POFs doped with conjugated oligofluorenes can act as optical amplifiers; an important component in the development of short haul POF networks. We have shown that the oligomers can withstand the POF fabrication procedure and that they are miscible enough in PMMA to produce films and POFs containing mainly isolated and non-interacting molecules. This is an exciting result as it paves the way for all optical ultrafast gain switching in POFs.

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