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ARTICLE *in* CHEMICAL PHYSICS LETTERS · OCTOBER 1999

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The dynamics of gain-narrowing in a ladder-type π -conjugated polymer

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Received 28 May 1999; in final form 20 August 1999

Abstract

Amplified spontaneous emission (ASE) indicated by photoluminescence spectral-narrowing is observed from a ladder-type conjugated polymer at high excitation densities. The narrowed emission shows a distinct blue-shift with respect to the photoluminescence emitted at lower densities. The line-narrowing is examined in detail using picosecond luminescence spectroscopy in conjunction with femtosecond pump–probe spectroscopy. The experiments give insight into the fundamental processes that lead to optical gain in conjugated polymers. Our results provide conclusive evidence that gain-narrowing occurs simultaneously with an energetic relaxation of localized excitations. The spectral properties of the ASE are closely linked to the dynamics of the excited states. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

The question whether it is more appropriate to describe conjugated polymers in terms of a one-dimensional semiconductor band model like, e.g., the Su–Schrieffer–Heeger (SSH) Hamiltonian [1] or in terms of a disordered molecular solid [2] has been controversially discussed over the last decade [3]. Closely linked to this question is the fundamental problem of the primary photoexcited states in these materials. Over the last years there has been abundant evidence in favour of the molecular exciton model. Within this model, conjugated polymers are

considered to be a disordered array of fully conjugated subunits (i.e., the respective oligomers of varying lengths), separated by topological faults of the polymer chain. The primary photoexcitations are singlet Frenkel-like excitons localized on these conjugated subunits. The statistical variation of the effective conjugation lengths and the random spatial distribution of the polymer segments lead to an inhomogeneously broadened density of excited states. Transfer of excited states among the polymer segments leads to spectral relaxation within the density of excited states. This effect has been observed experimentally [4] and has also been reproduced in theoretical model calculations [5,6].

The nature and dynamics of photoexcited states is of great importance not only from a fundamental

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point of view, but also with respect to possible applications of conjugated polymers in optoelectronic devices like, e.g., light-emitting diodes (Ref. [7] and references therein). This is even more true for high-power devices such as lasers based on conjugated polymers. After the realization of a polymer light-emitting diode and the subsequent optimization of this kind of devices, it seemed to be straightforward to investigate whether laser emission can be achieved from these materials and if a conjugated polymer laser diode is feasible.

Stimulated emission has first been observed in pump–probe experiments [8–10]. Although no net optical gain was observed in these early measurements, they provided a first hint that optical gain in conjugated polymers might be feasible. Another indication of optical gain is spectral-narrowing of the emission, which has been observed in several conjugated polymers [11–15]. In the meantime, laser emission from conjugated polymers has been reported by several groups using different feedback structures such as planar and ring microcavities [16–18], macroscopic resonators [19–21] or distributed feedback structures [22–24].

However, the fundamental gain mechanism in conjugated polymers, i.e., the mechanism of the observed line-narrowing and the nature of the emitting species, is still a matter of discussion. Within the molecular model, a four-level mechanism similar to dye lasers [15,25,26] has been proposed. The line-narrowing observed in thin films of conjugated polymers can then be explained by amplified spontaneous emission (ASE) [14,27–30]: spontaneously emitted photons are waveguided in the polymer film and induce stimulated emission processes. The occurrence of ASE requires optical gain in the material. Other groups have proposed biexcitonic emission [12,13] or superfluorescence [31,32] as explanations for the observed line-narrowing effects.

Recently, the gain dynamics have been investigated experimentally and also modelled theoretically by several groups [28,33,34]. However, these groups employed time-resolved pump–probe measurements in conjunction with steady-state emission spectroscopy to investigate the dynamics of the optical gain. In pump–probe measurements, the stimulated emission processes indicated by a positive $-\Delta\alpha L$ signal (where $\Delta\alpha$ is the photoinduced change in the

absorption coefficient and L is the sample thickness) are induced by photons of the probe beam. Stimulated emission processes are merely used to detect the transient population of excited states. Hence, the dynamics of the excited states can be evaluated from pump–probe spectra. In the case of ASE, the excited-state dynamics is of course substantially influenced by the stimulated emission processes. The decay constants of the pump–probe signal are expected to coincide with those of the emission if one assumes only one species of excited states. However, the spectral features of ASE such as line-narrowing cannot be observed in pump–probe measurements since spontaneously emitted photons are not detected in these experiments. This is also true for spontaneous emission that is subsequently amplified by stimulated emission processes. Therefore, in order to measure the dynamics of the line-narrowing, one has to perform time-resolved emission spectroscopy at high densities.

In this Letter, we present measurements of the spectrally narrowed emission with high temporal resolution. Hence, we are able to directly observe the interplay between the excited-state dynamics and the stimulated emission processes which lead to gain-narrowing. Our results will show that the observed line-narrowing can be completely explained within the molecular exciton model with no need to invoke further effects such as biexcitonic emission or superfluorescent emission.

2. Experimental

The polymer under investigation is a methyl-substituted ladder-type poly(paraphenylene) (Me-LPPP, inset of Fig. 1) [35]. The material is soluble and exhibits excellent film-forming properties combined with an extremely low defect concentration and a high photoluminescence (PL) quantum yield in the solid state. Unlike unsubstituted ladder-type poly(paraphenylenes), the methyl-substituted derivative shows only a low tendency to form aggregates which could act as traps for singlet excitations [10]. Films of high optical quality with a typical thickness of $L = 300$ nm were obtained by spin coating a solution of Me-LPPP in CHCl_3 onto quartz substrates. As the polymer film has an effective refractive index of

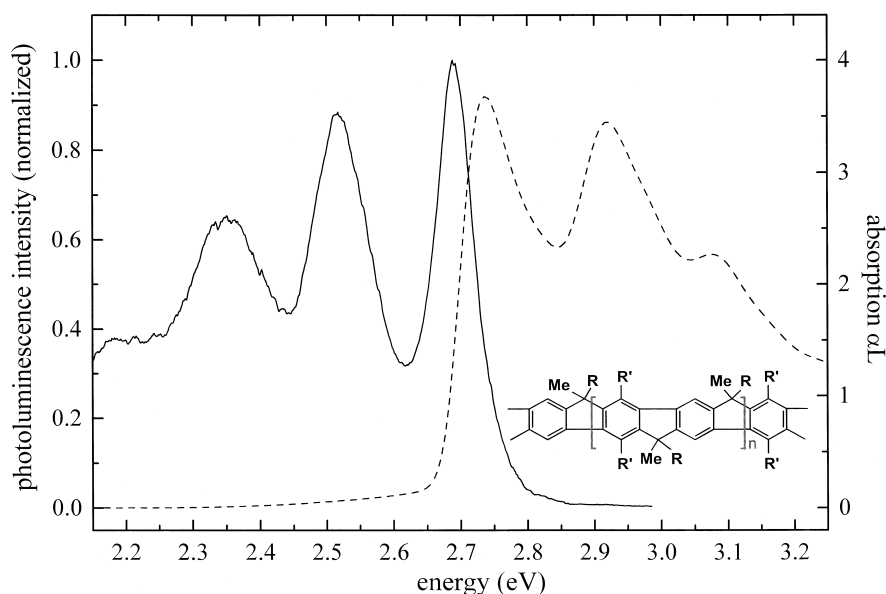


Fig. 1. Absorption (dashed line) and steady-state photoluminescence at low excitation density (solid line) of the Me-LPPP sample. The inset shows the chemical structure of Me-LPPP (R: $-1.4-\text{C}_6\text{H}_4-n-\text{C}_{10}\text{H}_{21}$; R': $-n-\text{C}_6\text{H}_{13}$).

about $n_{\text{eff}} = 1.66$, it forms an asymmetric waveguide structure with quartz ($n = 1.46$) on one side and air ($n = 1.0$) on the other side.

The steady-state emission spectra (PL and gain-narrowed emission; Figs. 1 and 2) were recorded using a tunable dye laser pumped by an excimer laser as excitation source. The dye laser delivered 20 ns pulses at a 10 Hz repetition rate. With pulse energies between 0.3 and 50 μJ focused onto a $7 \times 10^{-4} \text{ cm}^2$ spot, we achieved pump fluences up to 70 mJ/cm^2 . The emission was detected in backscattering geometry using a 0.27 m monochromator in conjunction with a liquid nitrogen cooled optical multichannel analyzer system.

In the time-resolved experiments (pump-probe and transient emission (Figs. 3–5)), we used the frequency-doubled output of a regeneratively amplified Ti:Sapphire laser system to excite the samples. The excitation pulses at a photon energy of 3.1 eV had a width of 200 fs at 1 kHz repetition rate. Excitation pulse energies were varied in the range from 0.1 to 100 nJ per pulse. In the pump-probe measurements the pump pulses were focused to a spot with a diameter of 40 μm , corresponding to a pump fluence of 4 mJ/cm^2 . A small fraction of the amplifier output was focused onto a sapphire crystal

to generate a spectrally broad white-light continuum, which was spatially and temporally overlapped with the pump-pulse on the sample to detect the photoinduced absorption change $-\Delta\alpha L$. The measured differential absorption spectra were corrected for the chirp of the probe pulse. In the transient emission experiments, the excitation pulses were focused to a line focus with a width of 100 μm and a length of about 300 μm at the sample edge leading to pump fluences between 1 and 15 $\mu\text{J}/\text{cm}^2$. The edge emission was dispersed in a 0.32 m monochromator and detected with a time resolution of 2 ps using a streakcamera setup. All experiments were performed at 300 K.

3. Results and discussion

Absorption and steady-state PL spectra at low excitation density (Fig. 1) show a pronounced vibronic structure. Both spectra can be explained assuming an inhomogeneously broadened density of excited states coupled to molecular vibrations. The PL spectrum is almost a mirror image of the absorption spectrum as required by the Franck–Condon principle. However, the spectral linewidth is narrower in the emission spectrum due to spectral relax-

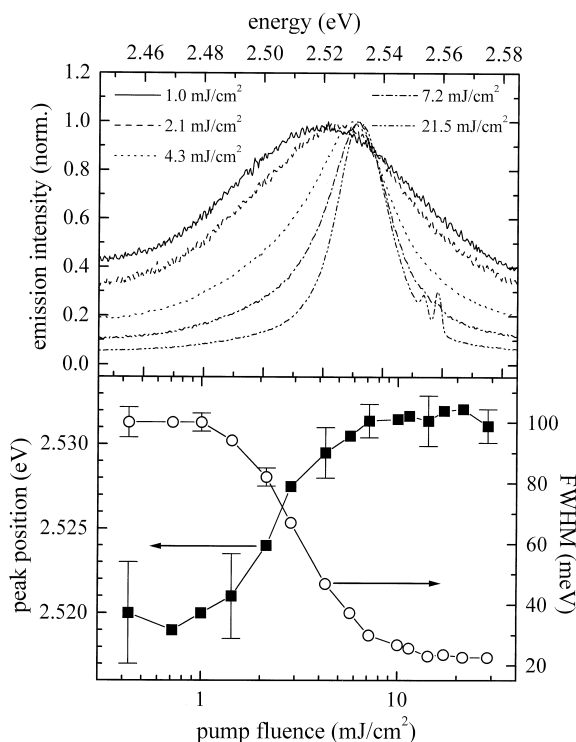


Fig. 2. Top: Evolution of the $S_1 \rightarrow S_0(0-1)$ emission band with increasing excitation density. The excitation photon energy is 2.75 eV. Bottom: Linewidth (FWHM, open circles) and peak position (solid squares) of the emitted spectra as a function of the pump fluence per 20 ns pulse.

ation of the excited states. The $S_1 \leftarrow S_0(0-0)$ transition is visible in the absorption spectrum at 2.74 eV followed by its vibronic progressions at higher energies. The PL spectrum shows the $S_1 \rightarrow S_0(0-0)$ transition at 2.69 eV followed by its dominant vibronic satellite around 2.52 eV. Absorption and PL spectra overlap in the region of their (0-0) transitions despite a shift of 50 meV due to spectral and structural relaxation. Hence, reabsorption of emitted photons is likely to occur, preventing the polymer from exhibiting optical gain in this spectral region. Therefore, we will concentrate on the $S_1 \rightarrow S_0(0-1)$ transition around 2.52 eV. Here, the absorption is very low allowing for optical net gain. In the following, this vibronic band will be termed the (0-1) transition, although it is in fact a superposition of several vibronic lines [25]. However, these closely spaced vibronic lines are usually masked by the inhomogeneous broadening, rendering only one vibronic band visible.

The evolution of the (0-1) emission band with increasing excitation intensity is depicted in the upper part of Fig. 2. The line width (full width at half maximum, FWHM) drops from 100 meV for the PL at low densities to 23 meV at high densities. Simultaneously, the emission peak shifts by 12 meV to higher energies, from 2.520 to 2.532 eV (Fig. 2, lower part). Line-narrowing occurs above a threshold pump fluence of about 1.5 mJ/cm^2 per 20 ns pulse. As already stated above, the line-narrowing can be explained in terms of ASE, indicating optical gain in the polymer material.

Differential absorption spectra show a positive $-\Delta\alpha L$ signal in the spectral region energetically above 2.30 eV (Fig. 3). At energies below 2.30 eV, we observe photoinduced absorption, most likely due to spatially separated excitons [8,10]. The minimum around 2.65 eV is most probably a photoinduced absorption band due to energetically higher excited states, superimposed on the positive signal. At even

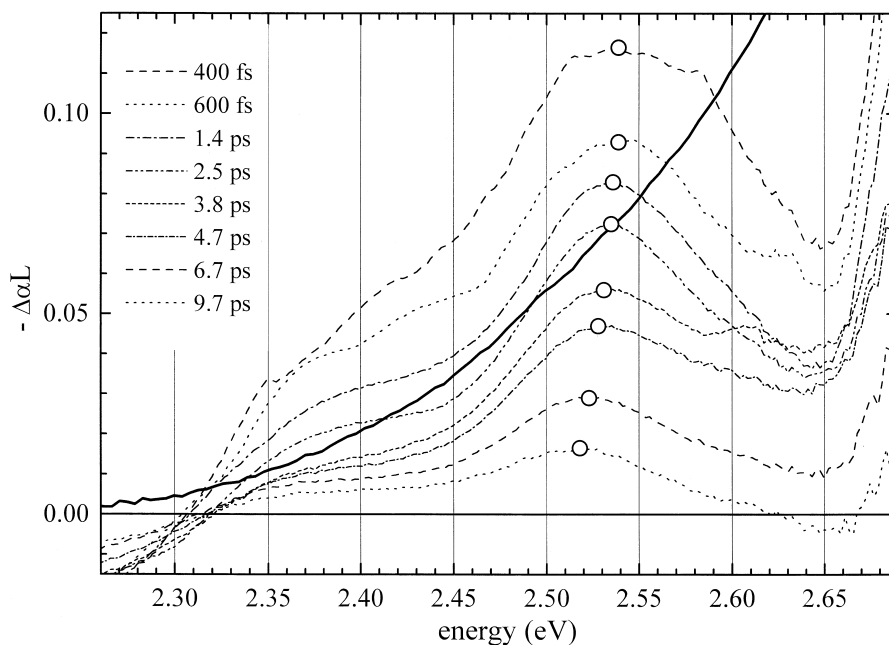


Fig. 3. Temporal evolution of the differential absorption spectra within the first 10 ps after excitation at 3.1 eV (dashed lines). The positive $-\Delta\alpha L$ signal between 2.30 and 2.65 eV is due to stimulated emission processes induced by the white-light probe beam. The circles indicate the maximum of the (0–1) stimulated emission band. The solid line shows the linear absorption αL .

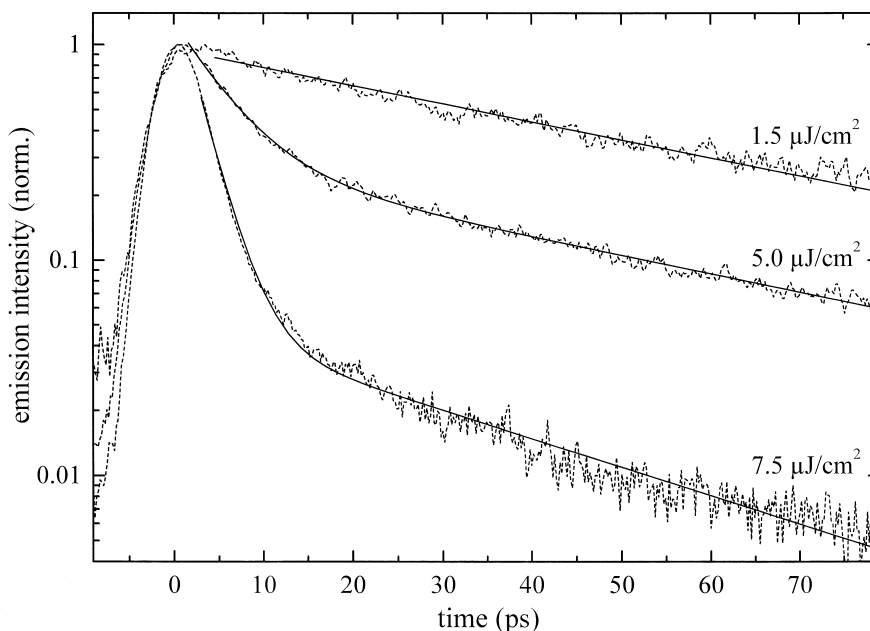


Fig. 4. Emission transients at the spectral position where gain-narrowing occurs (2.53 eV) for three different pump fluences on a logarithmic scale (dashed lines). The solid lines present single-exponential ($1.5 \mu\text{J}/\text{cm}^2$) and double-exponential (5.0 and $7.5 \mu\text{J}/\text{cm}^2$) fits to the measured transients.

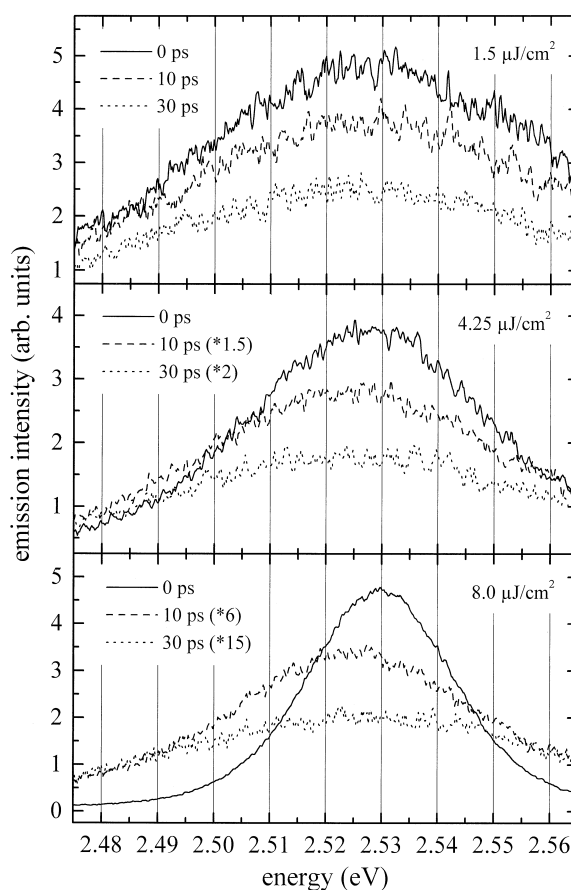


Fig. 5. Transient emission spectra at 0 ps (solid lines), 10 ps (dashed lines) and 30 ps (dotted lines) after excitation for three different pump fluences: $1.5 \mu\text{J}/\text{cm}^2$ (top), $4.25 \mu\text{J}/\text{cm}^2$ (center), $8.0 \mu\text{J}/\text{cm}^2$ (bottom).

higher energies, a large positive signal due to bleaching of the (0–0) absorption transition is visible. Here, we focus on the positive signal between 2.30 and 2.60 eV which is due to stimulated emission from the excited singlet states, induced by photons of the probe beam. The maximum between 2.50 and 2.60 eV maps the (0–1) emission. During the first few picoseconds after excitation, the maximum is blue-shifted with respect to the steady-state photoluminescence: the signal is due to excited states prior to spectral relaxation within the inhomogeneously broadened density of states. Spectral relaxation is clearly visible (circles in Fig. 3): the maximum of the positive $-\Delta\alpha L$ signal shifts by 20 meV from 2.539 eV (at a time delay of 400 fs) to 2.519 eV (at a delay of 10 ps). Spectral relaxation of excited states

has been observed experimentally in other polymers [4] and also in Me-LPPP [33] before. The effect has been modelled theoretically within the framework of the molecular exciton model as well [5,6].

It is important to note that the positive $-\Delta\alpha L$ signal must not be taken for optical gain. Gain occurs only if the photoinduced absorption change $-\Delta\alpha L$ is larger than the linear absorption αL . The gain coefficient g can be calculated as $g = (-\Delta\alpha L - \alpha L)/L$. In Fig. 3, the linear absorption αL is shown for comparison. It has been corrected for losses due to surface reflection and scattering at sample inhomogeneities, assuming that it approaches zero far below the absorption edge. The maximum gain can be found in the region between 2.515 and 2.530 eV; it reaches a value on the order of 2×10^3

cm^{-1} at a time delay of 400 fs after excitation. The positive $-\Delta\alpha L$ signal decays with a time constant of about 5 ps, dropping below the linear absorption 3 ps after excitation. Hence we can conclude that most of the optical gain in the Me-LPPP sample decays before the spectral relaxation of the excited states is completed.¹

However, as we have already pointed out before, the spectral features of ASE cannot be detected in pump–probe experiments. Therefore we performed time-resolved emission experiments, both in the low-density (PL) and the high-density (ASE) regime. Emission transients (Fig. 4), recorded at the maximum of the (0–1) emission (around 2.53 eV), show a strong dependence on the pump intensity. For low pump fluences ($1.5 \mu\text{J}/\text{cm}^2$), the transient can be fitted by a single exponential decay with a time constant of 50 ps (solid line in Fig. 4). For pump fluences above $2 \mu\text{J}/\text{cm}^2$ the decay becomes biexponential with fast decay constants in the range from 2.6 to 5 ps and slow decay constants between 35 and 50 ps. Both decay constants become faster upon increasing the pump fluences. The relative amplitude of the fast component grows from 0% (at pump fluences below $2 \mu\text{J}/\text{cm}^2$) to more than 95% (above $7 \mu\text{J}/\text{cm}^2$). Note that the fast component appears only above a threshold of about $2 \mu\text{J}/\text{cm}^2$ per 200 fs pulse, corresponding to an excitation density on the order of $1 \times 10^{17} \text{ cm}^{-3}$. The threshold for line-narrowing in the steady-state experiments, using 20 ns laser pulses to excite the sample, was at $1.5 \text{ mJ}/\text{cm}^2$ (Fig. 2). Taking into account the decay time of the fast emission component of about 4 ps, this corresponds to an effective excitation density on the

order of $5 \times 10^{16} \text{ cm}^{-3}$. Hence, the fast component in the emission transients occurs at nearly the same threshold excitation density where line-narrowing is observed in the steady-state experiments. The decay time of the fast component is approximately the same as the one observed for the decay of the positive $-\Delta\alpha L$ signal in the pump–probe experiments.

We have to note that we observed signs of sample degradation in the course of the time-resolved emission measurements, which were – in contrast to all other experiments – performed under ambient conditions. Especially the fact that the decay time of the slow (PL) component of the emission drops from 50 to about 35 ps at high pump fluences could be attributed to additional trapping in defect states generated by the high-intensity excitation light.

The observed emission dynamics is consistent with rate equation models incorporating stimulated emission at high excitation densities [28,29,34]: in these calculations, rate equations are used to model transient pump–probe data, i.e., the decay of a positive $-\Delta\alpha L$ signal. Hence, the models explain the temporal dynamics of the excited states in the presence of ASE, but not the spectral features of the emission, i.e., the line-narrowing and the blue-shift of the narrow emission line.

Time-resolved emission spectra for three different pump fluences are shown in Fig. 5. At a pump fluence of $1.5 \mu\text{J}/\text{cm}^2$ (top of Fig. 5), the emission maximum at 0 ps is at 2.527 eV. A detailed investigation reveals a slight spectral relaxation of about 2 meV to 2.525 eV within the first 30 ps. Increasing the pump fluence leads to a significant narrowing of the spectra at 0 ps, from a FWHM of 80 meV for low pump fluences to 31 meV at high pump fluences. Simultaneously, the emission peak at 0 ps shifts from 2.527 eV at low pump fluences to 2.530 eV at high pump fluences. This can easily be explained by the fact that spectral relaxation occurs already within the temporal resolution of 2 ps in the emission experiments (see also Fig. 3). The spectra at 30 ps, however, remain almost unaffected upon increasing the pump fluence: the peak position remains at 2.525 eV, the FWHM is in all cases on the order of 85 meV. A spectral relaxation, indicated by a dynamic shift of the emission maximum of 5 meV (from 2.530 eV at 0 ps to 2.525 eV at 30 ps), is

¹ One remark should be made about bleaching of the linear absorption αL (Fig. 3, solid line) which could, in principle, contribute to the positive $-\Delta\alpha L$ signal between 2.30 and 2.60 eV as well. The linear absorption in this spectral region below the (0–0) absorption band is either due to extremely low-lying bulk polymer states or – more probable – to a low concentration of aggregate states [10]. In either case, since these states emit at even lower energies, they will not take part in the stimulated emission processes which lead to ASE. Their lifetime is even longer than the lifetime of bulk polymer states near the center of the density of states [10] which is on the order of 50 ps (Fig. 4). Hence, it seems rather unlikely that these low-lying states influence the (sub-)picosecond dynamics observed here.

clearly visible at high pump fluences ($8.0 \mu\text{J}/\text{cm}^2$, bottom of Fig. 5). Additionally, the relative weight of the 0 ps spectra increases with growing pump fluences: at $1.5 \mu\text{J}/\text{cm}^2$ the maximum of the 0 ps spectrum is about 2 times larger than the spectrum at 30 ps; at a pump fluence of $8.0 \mu\text{J}/\text{cm}^2$ the ratio between the 0 ps spectrum and the 30 ps spectrum is about 35.

The time-resolved emission spectra clearly indicate that the gain-narrowed emission occurs only at high densities in the first few picoseconds after excitation. The narrow emission is obviously linked to the fast decay component observed in the emission transients. Simultaneously with the decay of the fast emission component, the spectra broaden and show a dynamic red-shift until they match the PL spectrum at low excitation densities. Conclusively, we can explain the spectra by ASE, i.e., spontaneously emitted photons are amplified by stimulated emission processes in the pumped region of the polymer. Amplified spontaneous emission requires optical gain and therefore a population inversion between the excited state and the ground state in the material. The high excited-state density required for a population inversion exists only for a few picoseconds and is rapidly reduced by stimulated emission processes. Spectral relaxation of the excited states as observed in pump–probe experiments occurs on the same timescale. As ASE is emitted from excited states prior to or simultaneously with their spectral relaxation, it must obviously be blue-shifted with respect to the steady-state PL spectrum at low excitation density which is mainly emitted from relaxed states. Once the density of excited states drops below the inversion threshold, the ASE process stops and the remaining excited-states decay radiatively emitting ‘usual’ PL. This is also supported by the fact that the slow decay constant of the transient emission (Fig. 4) at high excitation intensities is about the same as the one for the low-density PL. Again, this explanation is generally consistent with the models published by other authors [28,29,34]. However, our measurements show that it is necessary to incorporate the excited-state dynamics [5,6] into the models to fully explain the temporal and spectral behavior of the ASE. In Ref. [34] the authors argue that ASE is emitted *after* an energetic relaxation of the excited states has occurred. While this might be the case for

ultrafast vibrational relaxation processes which occur on a timescale below 200 fs, we see clear indications in our measurements that ASE occurs mainly *prior* to the spectral relaxation within the inhomogeneously broadened density of states.

4. Conclusion

The gain-narrowed emission we observed in a thin film of a ladder-type conjugated polymer can easily be explained within the molecular model for conjugated polymers, assuming singlet excitons as the only molecular excited states involved in the gain process. The blue-shift of the ASE with respect to the PL at low excitation densities can be attributed to the fact that ASE is predominantly emitted before spectral relaxation of the excited states within the inhomogeneously broadened density of states takes place. PL at lower densities on the other hand is mainly emitted after the relaxation process is completed.

ASE and spectral relaxation both occur on the same timescale of only a few picoseconds after excitation. Hence, the spectral position of the ASE is influenced by the excited-state dynamics. Although we cannot exclude the participation of other effects such as superfluorescence [31,32] or biexcitonic emission [13] in the line-narrowing process, based on our measurements we do not see any reason to invoke these effects in order to explain the emission-narrowing in conjugated polymers.

Acknowledgements

We gratefully acknowledge stimulating discussions with H. Bässler and W.W. Rühle. This work was supported by the Deutsche Forschungsgemeinschaft (Sonderforschungsbereich 383), the Stiftung Volkswagenwerk and the Fonds der Chemischen Industrie.

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