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Temporal behaviors of the amplified spontaneous emission and the lasing from a copolymer

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Abstract

The time and intensity dependent characteristics of the amplified spontaneous emission (ASE) and the lasing of a copolymer in tetrahydrofuran (THF) solution, Poly[1,8–octanedioxy–2,6–dimethoxy–1,4–phenylene–1,2–ethenylene–1,4–phenylene–1,2–ethenylene–3,5–dimethoxy–1,4–phenylene](CNMBC-Ph), were studied. By using Nd:YAG laser-streak camera system, we investigated the time behaviors of the ASE and the lasing of CNMBC-Ph pumped at various energies on picosecond time scale. Their real-time evolution of intensity-dependency reveals that the threshold and the time of the lasing generation are closely related to the temporal intensity of the pump light. The relationship between the threshold of pump energy and the duration of the pump light, spontaneous and the cross section of the stimulated emission were discussed.

Keywords: Copolymer; Lasing; Amplified spontaneous emission; Threshold; Spontaneous lifetime

1. Instruction

Since the advent of photopumped plastic lasers, conjugated polymers have become more attractive materials for applications in optoelectronic devices, and great interest in achieving lasing in conjugated polymers with electrical pumping is stimulated. The properties of excitation intensity dependent phenomena including line-narrowing, amplified spontaneous emission (ASE) and lasing in several polymer materials excited by femtosecond, picosecond and nanosecond pulses have been reported [1–11]. Some common properties of these polymers are high gain and absorption coefficient, large stimulated emission cross section and Stokes shift, low threshold and dramatic spectral narrowing; but the threshold of pump intensity is different because measurements have been performed in different materials, device configurations and excitation conditions.

In this work, we studied the time and intensity dependent properties of the ASE and the lasing of a copolymer, Poly[1,8–octanedioxy–2,6–dimethoxy–1,4–phenylene–1,2–ethenylene–1,4–phenylene–1,2–ethenylene–3,5–dimethoxy–1,4–henylene](CNMBC-Ph) (Fig. 1), in tetrahydrofuran (THF) excited by 355 nm picosecond pulses. The relationship between the threshold and the occurring time of the lasing, and that between the ASE of the CNMBC-Ph and the intensity and the shape of the pump pulses were investigated on the basis of the temporal evolution of the lasing and the ASE under different pump intensities. The stimulated emission cross section was estimated to be 2.19×10^{-16} cm^2. The influence of the spontaneous lifetime, stimulated emission cross section and the duration and rise-time of pump pulses on threshold energy of lasing was discussed.

2. Experimental methods

3 mg of CNMBC-Ph powder dissolved in 1.0 ml redistilled THF solution, and a quartz cuvette of 1×1 cm was used as a sample cell in experiments. All experiments were carried out in air at room temperature.

355 nm picosecond pulses from an active mode-locked Nd:YAG laser, operated at tripled frequency ($\lambda = 355$ nm, pulse width = 300 ps) with a repetition rate of 3 Hz were employed as the pump source. The pump beam was shaped by a cylindrical lens into a stripe of 200 μm in width and 5 mm in length as it was focused on the sample cell. Temporal evolution of ASE of the CNMBC-Ph was examined from an end of the stripe by a streak camera (M1763, Japan) with 10-picosecond time resolution.

The resonator of the polymer laser was made up of two flat mirrors, one of which was a high reflector (HR) and the

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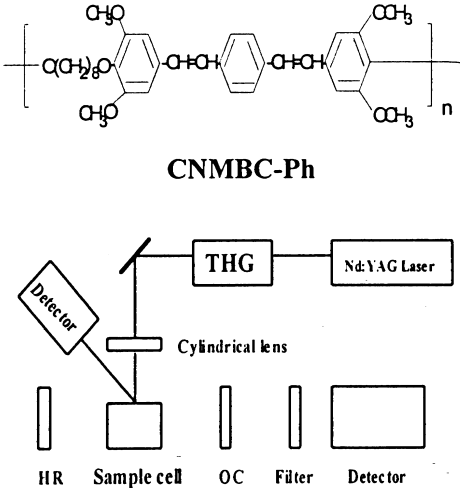

Fig. 1. Chemical structure of CNMBC-Ph and the experimental system.

other was an output coupler (OC) of a polished quartz flat, located at the two sides of the cell. The distance between the two mirrors is 14.5 mm. Therefore the optical length of the cavity is 20.5 mm. Temporal evolution of the laser pulse of the CNMBC-Ph solution pumped transversely by the stripe beam was also recorded by the streak camera. The double frequency pulses ($\lambda = 532$ nm) were used as reference pulses for the measurement of the temporal evolution of the ASE and laser pulses and of the fluorescence decay. Both the chemical structure of CNMBC-Ph and the experimental system are shown in Fig. 1.

3. Results and discussions

Fig. 2 gives the laser and the ASE Spectra of the polymer sample that were obtained under the same pump energy. Both of the spectra have the similar peak at 456 nm. The spectral linewidth of the ASE (dash line) is broader than that of the laser; this may result from the longitudinal mode selection in the laser cavity. The temporal evolution of the ASE and the laser under the same energy of 25 μJ is shown

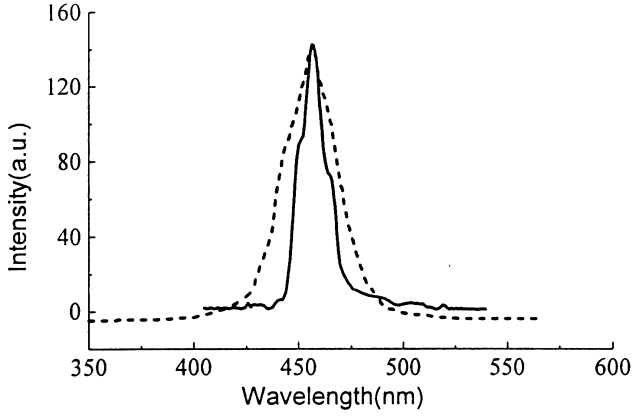

Fig. 2. The laser (solid line) and the ASE (dash line) spectra of the CNMBC-Ph in THF solution.

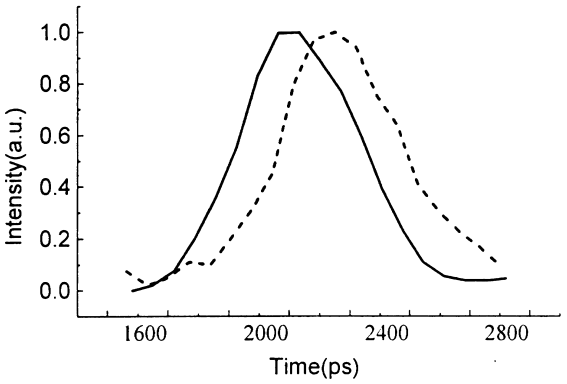

Fig. 3. Temporal evolution of the ASE (dash line) and the laser (solid line) under the same intensity of 25 μJ.

in Fig. 3. The laser pulse occurs about 135 ps ahead of the ASE pulse. It suggests that the light emission obtained with high gain during oscillating in the cavity occurs so quickly that it reaches the threshold before the ASE which is mirrorless [12,13]. In this case, the interval between the laser and the ASE is determined by the cavity losses. From Fig. 3, we can also calculate that the average lifetime of photons in the cavity is about 300 ps. From $\delta = L/\tau_R c$; where τ_R is the average lifetime of the photons in the cavity, $L = 20.5$ mm is the optical length of the cavity, c the velocity of

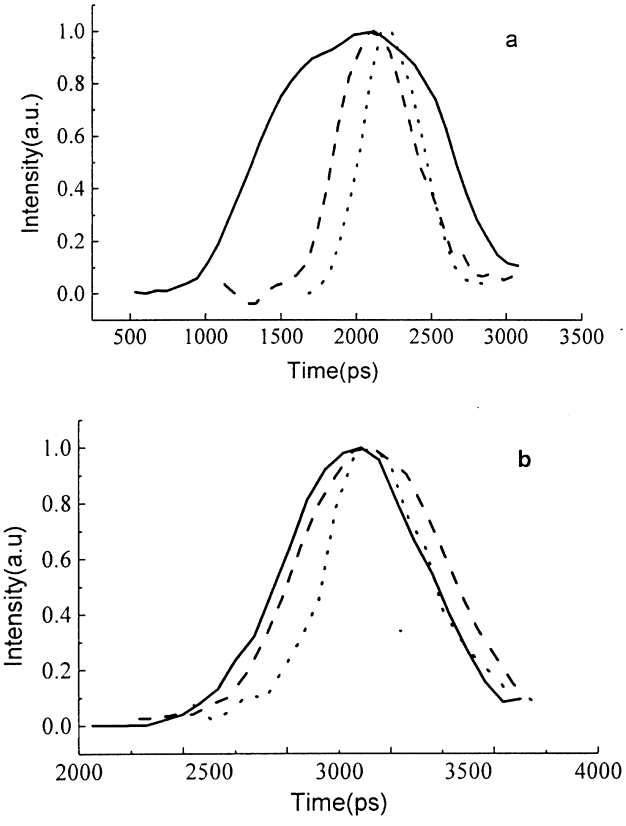

Fig. 4. The temporal evolution of the laser (a) and the ASE (b) under different pump energy: (1) 25 μJ (dotted line); (2) 40 μJ (dashed line); (3) 80 μJ (solid line).

light in vacuum, δ the factor of the total losses in the cavity, we get $\delta = 0.23$.

In inhomogeneous broadening active media, stimulated emission cross section can be estimated as [14]

$$\sigma_{SE} = \frac{\sqrt{\ln 2}\nu^2 A_{21}}{4\pi^{3/2}\nu_0^2 \Delta\nu} \quad (1)$$

where ν is the velocity of light in media, ν_0 resonance frequency, $\Delta\nu$ linewidth of inhomogeneous broadening, $A_{21} = 1/\tau_{21}$ the spontaneous emission rate and τ_{21} the average lifetime of excited state. Using Eq. (1) we obtained $\sigma_{SE} = 2.19 \times 10^{-16}$ cm^2, with $\tau_{21} = 1.2 \pm 0.2$ ns [15]. Obviously, Compared with the stimulated emission cross section of usually used laser gain media, which lies generally in the range of 10^{-18}–10^{-20} cm^2, the obtained cross section (σ_{SE}) is several orders of magnitude greater. It reveals that CNMBC-Ph polymer can fulfil lasing condition when the length of the active material is very short, or the cavity losses are relatively large.

Fig. 4 presents temporal evolution of the lasing and the ASE, which is calibrated by the reference pulses, the center of which is defined as the time start, under different pump energy. It shows that when the pump energy is raised, the laser and the ASE occur at an earlier time. Because the duration of the pump pulse is much shorter than the photoluminescence lifetime of CNMBC-Ph in solution, the polymer laser works in unsteady state. The effect of the spontaneous and non-radiative transitions from the upper level of the laser on population inversion can be neglected during the time of the excitation pulse. In this case, the threshold energy of pump pulse (E_{pt}) can be [14]; $E_{pt} = h\nu_p \delta V/\eta\sigma_{21} l$. Where ν_p is the frequency of pump light, η quantum efficiency, V volume of active gain media, l length of active gain media and δ losses. Therefore, cumulation of the upper level is directly related to the temporal photons or the temporal profile of the pump pulse. The occurring time and the pulse shape of the laser and the ASE are effected by both the pump intensity and the shape of the pump pulse.

The time behavior of the polymer lasers is similar to that of the conventional dyes. Both have spontaneous emission lifetime in nanosecond range and four-level system. If a polymer laser is pumped by a giant pulse where peak power is slightly above the threshold, then the polymer laser emits a lower pulse of duration approximately equal to the time that the pump pulse remains above threshold. Increasing the peak power of the pump pulse has the consequence that threshold is reached at an earlier time during the rise of the pump pulse. The shape of the polymer laser pulse then follows more or less the pump pulse because of the short lifetime of the polymer and, in most cases also, of the cavity. Owing to short spontaneous lifetime of conjugated polymers, increasing the duration of the pump pulse may increase the spontaneous emission loss of the gain media. Thus the threshold energy of the pump pulse may increase as the pulse duration is prolonged, and the pump pulse energy may not reach the threshold of lasing. In conclusion, the shape of the polymer laser pulse is closely related to that of the pump pulse if pump pulse duration is shorter than or equivalent to the spontaneous lifetime of the polymer.

Because the ultrafast pulse lasers have been widely used as pump sources of conventional dye lasers, the photopumped conjugated polymer lasers with various configurations such as microcavities, distributed feedback resonators and external resonators, have broad prospects for replacing conventional dye lasers, especially those in the blue wavelength region. However, on the basis of the above discussion, it is impractical to achieve electrical pumped conjugated polymer lasers only by increasing current densities to reach the threshold excitation density. Therefore, in order to realize electrically pumped conjugated polymer lasers, some other means such as selecting conjugated polymer with long spontaneous lifetime and employing suitable laser cavity configurations to increase gain also need to be considered.

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