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Cooperative and stimulated emission in poly(*p*-phenylene-vinylene) thin films and solutions

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We discuss cooperative and stimulated emissions and separate their respective contributions to the emission spectral narrowing in thin films and solutions of poly(*p*-phenylene-vinylene) derivatives. Whereas cooperative radiation is favored in films with poor optical confinement, directional stimulated emission is dominant in dilute solutions and thin films with superior optical confinement. Spectral narrowing in the latter case could be achieved by increasing either the excitation length or excitation intensity, from which we determined the optical gain and loss coefficients at the 0-1 emission band assuming a simple amplified spontaneous emission model. We found that the threshold excitation intensity for stimulated emission in neat films is mainly determined by self-absorption and that the gain saturates at emission intensities of about 10^7 W/cm^2. [S0163-1829(98)02015-3]

I. INTRODUCTION

Recent demonstration of dramatic emission spectral narrowing in poly(*p*-phenylene-vinylene) (PPV) (Refs. 1–4) and other π-conjugated polymers,[5–10] in the forms of solutions,[1,7] as well as thin solid films,[2–10] has stirred widespread interest to the phenomenon of laser action in the field of conducting polymers. Spectral narrowing in these materials occurs at relatively high excitation intensities and is typically accompanied by substantial excitonic lifetime shortening.[4,5] The origin of this spectral narrowing has been independently attributed to lasing,[1,2] amplified spontaneous emission (ASE),[3] and cooperative radiation or superfluorescence (SF).[4] High optical gain is required for all of these nonlinear optical phenomena. An additional requirement for lasing is the presence of optical feedback, which typically results in well-defined cavity-dependent laser modes. Such laser action has indeed been demonstrated in Fabry-Perot type resonators,[1,8] and also in planar[2] and cylindrical[11] microcavities. However, spectral narrowing has been observed also in thin π-conjugated polymer films, where the existence of an optical feedback mechanism necessary for lasing is not obvious. In this case explanations involving ''mirrorless lasing'' phenomena, such as ASE (Refs. 3,5–7) and SF (Refs. 4,7), have been evoked. Whereas both lasing and ASE are the direct result of stimulated emission (SE) processes, SF is a cooperative spontaneous process, which is due to the buildup of a macroscopic optical dipole moment ensuing from coherent interactions between the photogenerated excitons via their radiation electromagnetic field.[12–14]

Scientific interest toward the phenomenon of spectral narrowing in conducting polymers continues to grow. This is largely due to the possible application of these polymers as active laser media in future plastic laser diodes. Hence better understanding of the mechanisms leading to spectral narrowing and the criteria determining their respective contributions is needed. In the present studies we investigate ''mirrorless'' nonlinear emission in thin films and solutions of a soluble derivative of PPV, namely, 2,5-dioctyloxy PPV, or DOO-PPV.[4] We separate the respective contributions of SF and SE to the spectral narrowing and show that SF is dominant in thin films with poor optical confinement, whereas SE prevails in dilute solutions and thin films with superior optical confinement. In the latter case, by using an excitation area in the shape of a narrow stripe, we obtained directional SE in the form of a narrow waveguided beam inside the DOO-PPV films, which was achieved by increasing either the excitation length, or the excitation intensity. Using a simple model of a one-dimensional ASE process, we subsequently estimated the exponential gain coefficient for different excitation intensities at wavelengths corresponding to the 0-1 polymer emission band and estimated the optical loss coefficient in this spectral range to be about 30 cm^{-1}.

II. EXPERIMENT

DOO-PPV films with uniform thickness d ranging from 0.5 to 4 μm were spincast onto quartz substrates at spinning speeds from 100 to 2000 rpm from fresh, 5% by weight, chloroform solutions. The excitation source was the frequency-doubled output (at 532 nm) of a Nd:YAG regenerative laser amplifier producing ~100-ps pulses at 500 Hz. The pump laser beam was focused onto the polymer film or a square, $1\times1\times3$ cm cuvette containing the polymer solution in chloroform, using either a round lens (for round excitation area) or a cylindrical lens (for obtaining an excitation area in the form of a 100-μm-wide stripe). In the latter case the length of the rectangular excitation area could be varied from 100 μm to 6 mm using a variable-width slit in front of the polymer film or solution, which could block parts of the pump beam. The polymer emission was collected from either the front or the side of the substrate (or the cuvette) and spectrally analyzed using a 0.25-meter spectrometer, a Si photodiode and a lock-in amplifier (SR 850). The spectral resolution of the apparatus was 1.5 nm. All measurements were performed in a dynamic vacuum of 10^{-4} torr at room temperature.

III. COOPERATIVE EMISSION

A. Semiclassical model of superfluorescence

Substantial emission spectral narrowing may occur in some materials without a resonator, i.e., after a single pass through the medium with extremely high optical gain. This phenomenon has been dubbed mirrorless lasing,[15] which includes superradiance,[16] SF and ASE. The former two processes are examples of *cooperative* emission, whereas ASE is a *collective* process. At low densities following optical excitation, excitons or any other optical emitters decay via spontaneous emission. The individual emitters then act independently of the radiation field, their respective phases are completely random, and their emission is characterized by a radiative decay time τ_0. At high exciton densities, in case of high photoluminescence (PL) quantum yield and high optical gain, however, this simple behavior may change due to a strong interaction between the excitons through their own radiation field.[13,14] The decay of the excited state then occurs via SF in a much shorter characteristic time.

Superfluorescence (SF) is a cooperative spontaneous emission from an ensemble of electrical dipole emitters.[17] Depending on the emitter dynamics, this emission may have a high degree of coherence and therefore resemble laser emission. Following the original work of Dicke,[16] the effect has been theoretically studied in details.[12–14,17] However, its experimental observations in solid-state systems have been rare.[18–20] In SF, the initial ordering process among the emitters is induced by a common radiation field overlapping different emitters. This leads to the buildup of correlation among the dipole moments belonging to different emitters, and the ensemble of phase-locked emitters, each having a dipole moment μ, acquires a macroscopic dipole moment $N_c\mu$, where N_c is the number of correlated emitters.[12,16] This macroscopic dipole then radiates spontaneously in a well-defined direction depending on the geometry of the sample with a higher rate and much stronger peak intensity then the total emission of the independent dipoles. The *coherent* radiation power is proportional to $(N_c\mu)^2$ and the emission emerges in a short pulse with time duration of order τ_0/N_c.[16] In order to observe SF, the gain coefficient of the emitters must be large and the sample length must be small compared to the distance that the radiation can travel in the medium within the dipole dephasing time T_2^*. In this case radiation coming from any dipole can be strongly amplified and transmitted to another dipole and vice versa before any of these dipoles has either radiated spontaneously or been dephased.

The main features of SF, such as excitation intensity dependence, emission pulse shortening, time delay, etc., can be described within a simplified semiclassical approach, which uses Maxwell-Bloch equations while neglecting the dipole-dipole interaction.[13,14] It was shown by Bonifacio and Lugiato[13] that in a mean-field approximation the system of noninteracting emitters is described by a pendulum equation with two driving terms:

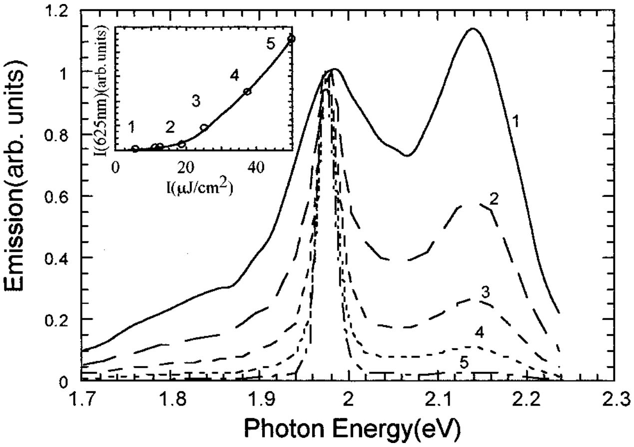

FIG. 1. Normalized emission spectra of a DOO-PPV film at various pulsed excitation fluence: $I_1=10$ μJ/cm^2, $I_2=2I_1(x1/3)$, $I_3=3I_1(x1/8)$, $I_4=5.4I_1(x1/26)$, $I_5=25I_1(x1/200)$. The inset shows the amplification at 625 nm close to the threshold intensity $I_0=I_2$.

$$\ddot{S}_Z+\left(\kappa+\frac{1}{2T_2^*}\right)\dot{S}_Z=-\frac{g_0^2}{V}\,e^{-t/T_2^*}(2S_T^2+4A_T^2S_Z), \qquad (1)$$

where S_Z is the exciton population, S_T describes the cooperative, macroscopic dipole moment of the system, A_T is the photon number operator of the emitted electromagnetic field, κ is the radiation leakage rate out of the active volume V, T_2^* is the inhomogeneous dephasing time, and g_0 is the coupling constant, which is proportional to the exciton oscillator strength.[13] It is apparent that the right-hand side of Eq. (1) contains two driving terms, which correspond to the macroscopic dipole moment of the excitons (S_T^2) and their radiated electromagnetic field (A_T^2), respectively. The first term gives rise to cooperative radiation or SF, whereas the second term is the source of SE. It thus becomes apparent that, in general, the resultant emission contains contributions from both SF and SE. Whether this nonlinear emission is dominated by SF or SE, is determined within this model by the values of $\kappa\tau_c$ and T_2^*. Here τ_c is the Arrechi-Courtens cooperation time[12] given by $(g_0\sqrt{N/V})^{-1}$, where N is the number of excitons. The value of $(\kappa\tau_c)^{-1}$ accounts for the number of photons emitted via the SE process.[21] Therefore, in the case of strong optical confinement, where $\kappa\tau_c<1$, SE is the dominant radiation process; on the other hand, for weak optical confinement, where $\kappa\tau_c>1$, cooperative emission is the primary process.[13] The latter also requires that $T_2^*>1/\kappa$, so that the case of ''pure'' SF can be identified by the following conditions:

$$1/\kappa<\tau_c<T_2^*. \qquad (2)$$

B. Spectral narrowing in films with poor optical confinement

Figure 1 illustrates the spectral narrowing process previously observed in thin DOO-PPV films using a round excitation area.[4] It is seen that the initially broad (~ 80 nm) PL emission spectrum collapses into a narrow spectral line of $\sim 7-8$ nm at ~ 630 nm for excitation intensities I exceeding a well-defined threshold excitation intensity I_0 of order 10 μJ/cm^2. The peak emission intensity dependence on I is

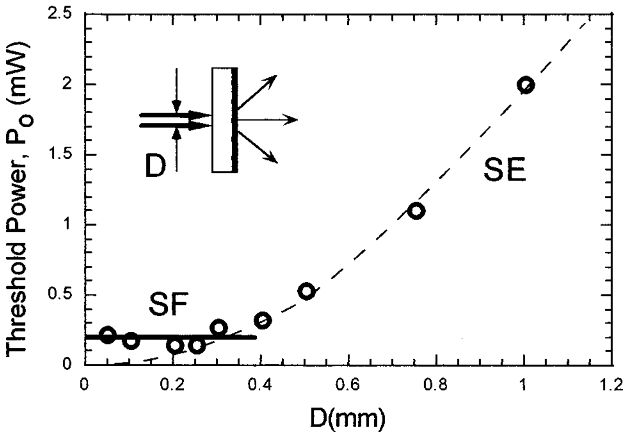

FIG. 2. The threshold excitation power P_0 for obtaining spectral narrowing in isotropically emitting DOO-PPV films at various diameters D; the full and the dashed lines through the data points are the expected dependencies for SF and SE, respectively (see the text). The inset illustrates the excitation setup, where D is the diameter of the round illuminated area.

superlinear for $I > I_0$, as shown in Fig. 1 inset; thus I_0 can be determined from the onset of this superlinear dependence at 630 nm.

Our previous measurements showed[4] that the emission from DOO-PPV films remained isotropic at all excitation intensities, and no well-defined waveguiding was observed. Similar results have been obtained using other luminescent π-conjugated polymer films.[22] This isotropic emission pattern could be attributed to poor film quality, which consequently leads to strong light scattering inside the polymer film and on its surface. Optical confinement in such films is poor and the characteristic length for the nonlinear emission process L_s is on the order of few d. On the other hand, a SE process requires a substantially longer characteristic length for appreciable amplification,[4] thus SF appears to be a more appropriate explanation for the isotropic narrow-band emission pattern. Instead of intensity I_0, the onset of spectral narrowing may be characterized by a threshold excitation power P_0. Using the same DOO-PPV films that were used before in Ref. 4, we measured the dependence of P_0 on the diameter D of the round excitation area, as shown in Fig. 2. It can be seen that P_0 is practically constant in the range of 50 μm$<D<$300 μm; as clarified below, this behavior cannot be explained by a simple SE process, such as ASE. Ignoring losses and assuming that light amplification occurs in the direction of maximum excitation length (i.e., D) parallel to the film surface, a simple SE process at threshold satisfies the relation $\gamma_0 D \sim 1$, where γ_0 is the threshold gain coefficient given by $N_0\sigma$, where N is the photogenerated exciton density and σ is the optical emission cross section. For intermediate excitation intensities, where the bimolecular recombination is negligibly small, $N_0 \sim P_0/V$ and $V = \pi D^2 d/4$, leading to $P_0 \sim \pi D d/4\sigma$ at threshold this functional dependence on D cannot explain the P_0 independence on D for D values up to 300 μm, as shown in Fig. 2. In the case where bimolecular recombination is important and occurs for times $t < t_p$ (t_p is the excitation pulsewidth), we get $N_0 \sim P_0{}^{1/2}/V$, which leads to $P_0 \sim D^2$ also in contrast with Fig. 2. Alternatively, if the characteristic length of the SE process is L_s and, as argued before, much smaller than D, then the threshold is mainly determined by self-absorption and scattering. From $\gamma_0 L_s \sim 1$ we similarly obtain in this case $P_0 \sim \pi D^2 d/4\sigma L_s$, a functional dependence on D, which is also inconsistent with Fig. 2 for D smaller than 300 μm.

On the other hand, a SF process is governed by the total *number* of photogenerated excitons and thus is determined by the excitation *power*,[12,17] which implies that P_0 remains constant and independent on D for excitation areas within the SF cooperation length.[13] SF is therefore a more plausible explanation of the P_0 independence on D at small values of D. From the length of the plateau in Fig. 2 we estimate the maximum cooperation length, l_c, of excitons in DOO-PPV films to be about 300 μm. In the semiclassical approximation l_c is given by $l_c = c\tau_c/2n_f$,[13] where n_f is the refraction index of the polymer film.[23] Moreover, the SF conditions (2) require that $T_2^* \geq \tau_c = 2n_f l_c/c$, from which we estimate a relaxed exciton dephasing time T_2^*, to be about 3 ps.[4] The right-hand side of condition (2) can also be satisfied for films with poor optical confinement, where we estimate $\kappa \sim c/n_f L_s \sim (10\text{–}100\ \text{fsec})^{-1}$. We therefore suggest that SF is dominant in these DOO-PPV films for $D<300$ μm (Fig. 2). However, for $D > l_c$, the SF conditions (2) are no longer satisfied, indicating that in this case SE is the primary emission process. Accordingly, P_0 dependence on D in Fig. 2 for $D>300$ μm may be approximated by $P_0 \sim D^2$.

IV. STIMULATED EMISSION

A. Spectral narrowing in dilute DOO-PPV solutions

SE should occur whenever conditions (2), namely, $1/\kappa < \tau_c < T_2^*$, are violated,[13,17] which can be achieved by either decreasing κ or increasing τ_c. Hence, by diluting the DOO-PPV chromophore concentration in solution (leading to smaller N/V), we are able to increase τ_c and subsequently suppress SF. Figure 3(a) shows the emission spectra of a dilute DOO-PPV solution in chloroform (concentration of $\sim$2 g/L), which were measured at various excitation intensities using transverse photoexcitation with an excitation area in the shape of a stripe, as shown in Fig. 3(a), inset. The cuvette with solution was tilted in order to avoid lasing due to the reflections off its sides. It can be seen that spectral narrowing here (Fig. 3) significantly differs from that in DOO-PPV films (Fig. 1). This includes the peak wavelength of 590 nm, rather than 630 nm in films, and the final linewidth of 20 nm instead of 7 nm in films. We also measured the dependence of the emission peak intensity I_{se} at 590 nm on the excitation intensity I as shown in Fig. 3(b). I_{se} grows exponentially with I, which is consistent with a simple ASE process[24] described by

$$I_{se} = \beta(e^{(\gamma-\alpha)L} - 1), \tag{3}$$

where β is a constant that depends on the excitation geometry,[24] L is the excitation stripe length, and γ and α are, respectively, the optical gain and loss coefficients at $\lambda \sim 590$ nm. Since γ is linear with I in the first approximation, then $\ln(I_{se}) \sim I$ at large values of I_{se}, in agreement with the

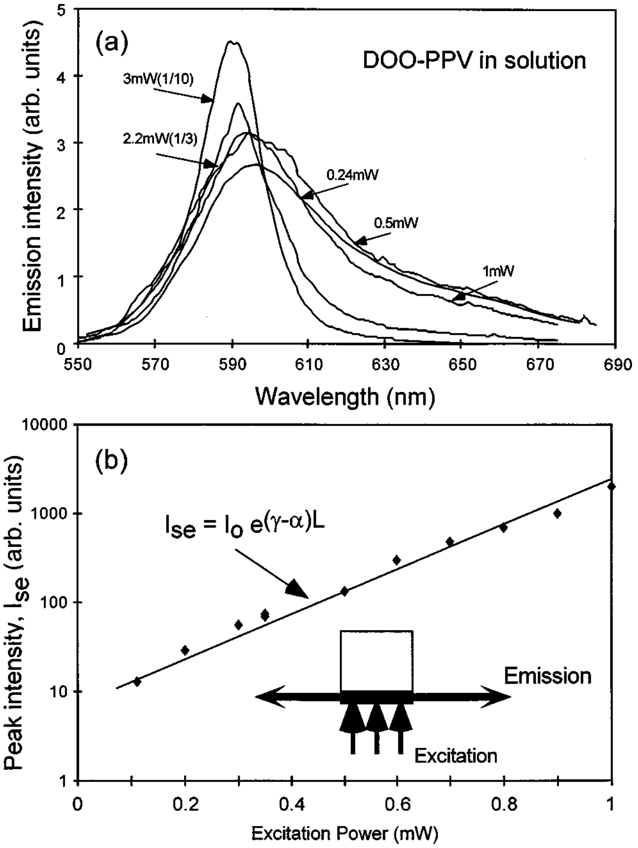

FIG. 3. Emission spectra in dilute DOO-PPV chloroform solutions at various excitation intensities I (a) and the dependence of the emission peak intensity at 590 nm on I (b); the line through the data points is a fit using ASE [Eq. (3)]. The inset shows the experimental setup for measuring the nonlinear emission from solutions.

data and fit shown in Fig. 3(b). These results show that indeed spectral narrowing in dilute DOO-PPV solutions is due to a single-pass ASE.

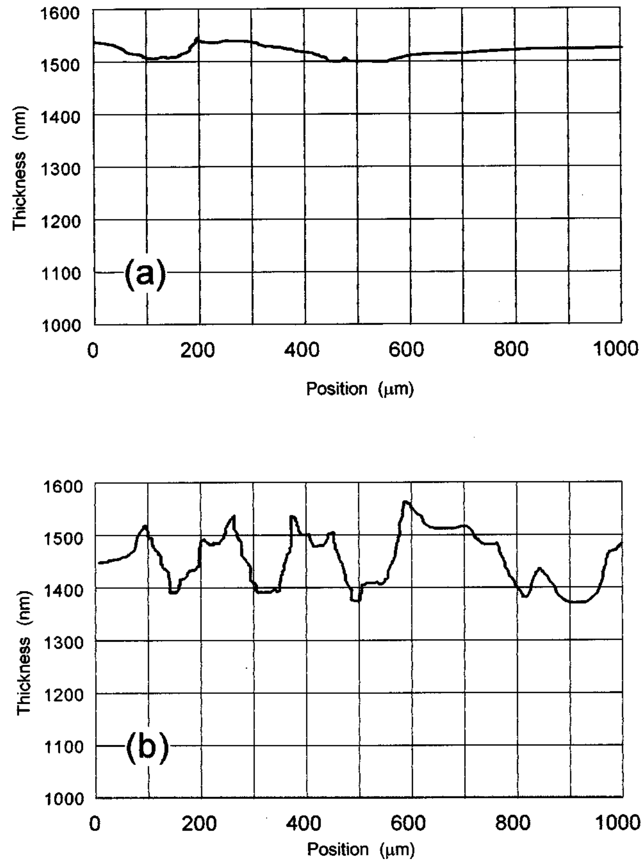

FIG. 4. Characteristic film thickness variation vs the film length for an improved DOO-PPV film (a) and a DOO-PPV film of inferior optical quality (b) used previously (Ref. 4).

B. Spectral narrowing in DOO-PPV films with superior optical confinement

Alternatively, we can achieve the condition $\kappa\tau_c \ll 1$ by decreasing κ. This, however, requires substantial improvement in the optical confinement properties of the polymer film. Recently, we have been able to improve the quality of our films using spinning speeds of 100–300 rpm, and thus spin-coated low scattering planar DOO-PPV waveguides, where a large portion of the polymer emission was optically confined inside the polymer film. Figure 4 compares the cross-sectional profile of such a superior DOO-PPV film (a) with that of the previously used DOO-PPV film (b). The film thickness was measured at different lateral positions using a Sloan Dektek profilometer. The improved DOO-PPV films are characterized by more uniform thickness with about 3% variation per 1 mm length, whereas the previously used films have thickness variations of 15–25 % per 1 mm length. The superior DOO-PPV films also show much better optical confinement, which in part may be explained by their better surface quality. Using the refraction index of the glass substrate, $n_s = 1.46$, and that of the DOO-PPV film, $n_f = 1.7$,[22] we estimate the maximum fraction f of emission which is waveguided or ''trapped'' inside the film[25] to be

$$f = \sqrt{1 - \left(\frac{n_s}{n_f}\right)^2} = 0.51.$$

As shown in Fig. 5, the spectrally narrow SE (bandwidth of ~8 nm) from such films was observed only in the direction parallel to the film surface, whereas the emission perpendicular to the film surface remained spectrally broad (~80 nm) even for $I > I_0$. This indicates that SE is enhanced due to waveguiding along the film, where the emission experiences the largest gain. Therefore, for the following measurements of the directional SE we used an excitation area in the shape of a narrow stripe (~100-μm wide). As a result, SE was predominantly emitted along the axis of the stripe, parallel to the film surface. The directional SE appeared in the form of a thin narrow beam propagating outside the excitation area, where it was scattered on the edge of the film. A part of this scattered light was trapped inside the quartz substrate; it was collected by a round lens in front of the monochromator and used for the spectral analysis of the DOO-PPV emission.

Figure 6 shows the directional SE spectra obtained by increasing either the excitation intensity I (a) or the excitation stripe length L (b). The results are virtually identical: in both cases spectral narrowing of the polymer emission was

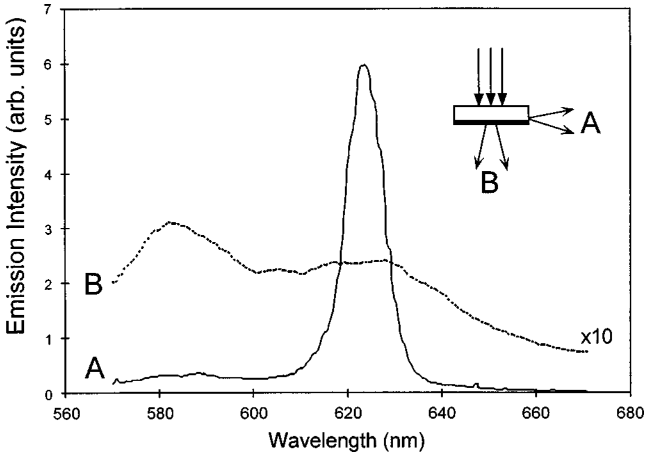

FIG. 5. The emission spectra of a superior DOO-PPV film measured in the directions parallel (A) and perpendicular (B) to the film surface, at the same excitation intensity, of about 1 MW/cm^2, note that the emission intensity B is magnified by 10. The inset illustrates the experimental setup.

observed above certain threshold values for both I and L. This directional SE can be successfully modeled using the ASE approximation and Eq. (3).[24] We assume that $\gamma \propto I$ and α is constant. Since γ has a maximum at $\lambda \sim 630$ nm, then $I_{se}(630\,\mathrm{nm})$ experiences the maximum gain, whereas amplification at other λ is relatively smaller. Consequently, this nonlinear amplification process leads to spectral narrowing when either I or L increases.

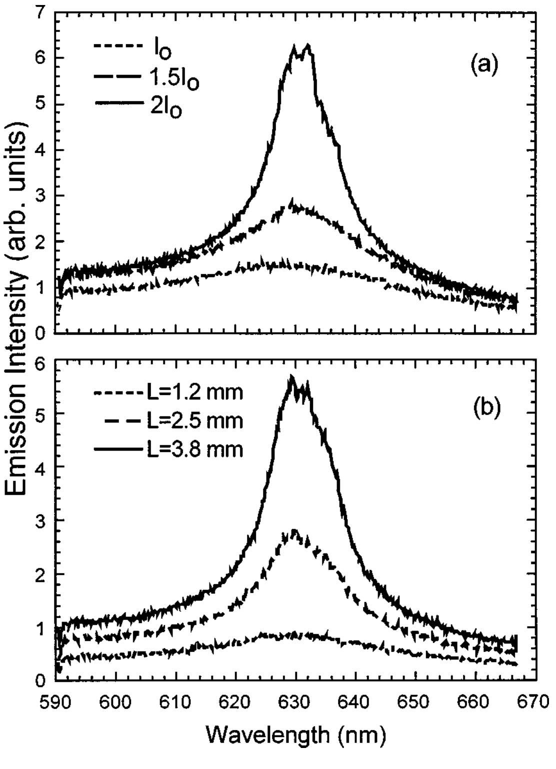

FIG. 6. Spectra of directional SE in superior DOO-PPV films obtained by increasing either the excitation intensity I (a) or excitation length L (b). The respective intensities I and lengths L are given, where $I_0 \sim 0.2$ MW/cm^2.

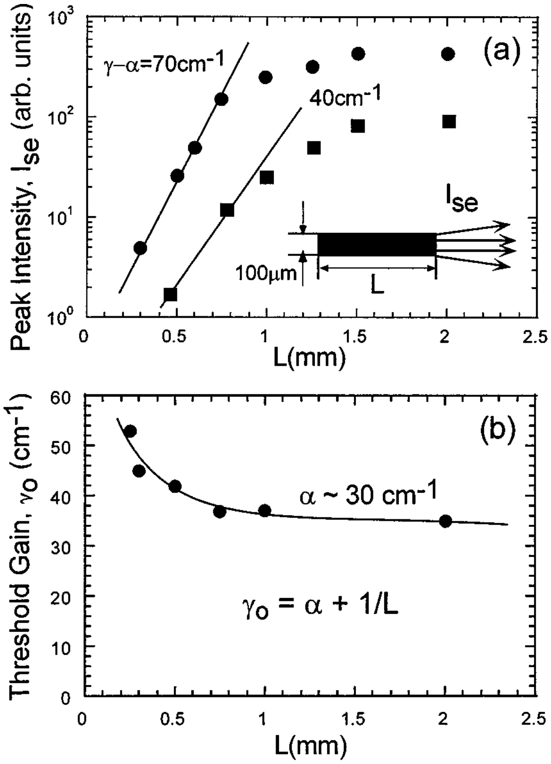

FIG. 7. (a) The peak emission intensity dependence on L for $I = 0.6$ MW/cm^2 (circles) and 0.4 MW/cm^2 (squares); the schematic illumination and collection methods are shown in the inset. (b) The SE threshold gain coefficient γ_0 obtained for various L. The lines through the data points are fits explained in the text.

We measured I_{se} dependence on L at different I, as shown in Fig. 7(a). In accordance with Eq. (3), SE grows exponentially at small L. This allows us to estimate the effective gain coefficients: $(\gamma - \alpha) \sim 70$ cm^{-1} for $I = 0.6$ MW/cm^2 ($N \sim 4 \times 10^{17}$ cm^{-3}) and ~ 40 cm^{-1} for $I = 0.4$ MW/cm^2 ($N \sim 2.5 \times 10^{17}$ cm^{-3}). From these measurements we obtain $\gamma \approx 170 \cdot I(\mathrm{MW/cm^2})$ (or $\gamma \approx N\sigma$, where $\sigma \sim 0.25 \times 10^{-15}$ cm^2) and $\alpha \sim 30\,\mathrm{cm}^{-1}$. In the ASE model, there exists a saturation emission intensity I_{sat} at which the optical gain saturates as I_{se} approaches I_{sat}. As a result, the emission rate at $\lambda = 630$ nm is approximately equal to the pump excitation rate at $\lambda = 532$ nm for $I_{se} \geqslant I_{sat}$; in the case of a loss-limited gain saturation, I_{se} completely stops growing.[24] From the onset of gain saturation in Fig. 7(a) (Ref. 26) we estimate that I_{sat} is of order 5×10^7 W/cm^2 or less. On the other hand, using $I_{sat} = hc/\lambda\sigma\tau$ (Ref. 24) (τ is the exciton lifetime), $\sigma \sim 0.25 \times 10^{-15}$ cm^2 and $\tau \sim 300$ ps,[4] we calculate I_{sat} to be of order 4×10^6 W/cm^2, which is consistent with our measurements.

We can define I_0 for SE by the onset of nonlinear amplification at 630 nm, which occurs when $(\gamma_0 - \alpha)L = 1$; this condition may be rewritten as follows:

$$\gamma_0 = \alpha + \frac{1}{L}. \qquad (4)$$

Figure 7(b) shows the threshold gain γ_0 measured at various L; γ_0 was calculated from the measured threshold intensity and the previously determined relation between I and γ.

TABLE I. Optical parameters for DOO-PPV films at 630 nm; l_c is the exciton cooperation length T_2^* is the relaxed exciton dephasing time, σ is the exciton SE optical cross section, α is the optical absorption coefficient, I_{sat} is the saturation emission intensity, and Q_{abs} is the self-absorption limited laser quality factor.

l_c	T_2^*	σ (630 nm)	α (630 nm)	I_{sat}	Q_{abs}
300 μm	$\geqslant 3$ ps	0.25×10^{-15} cm^2	30 cm^{-1}	$\sim 10^7$ W/cm^2	3000

Using Eq. (4) we can model the functional dependence in Fig. 7(b) and from the fit calculate α at $\lambda = 630$ nm to be $\sim 30\ \mathrm{cm}^{-1}$, in agreement with the above estimate from the relation between I and γ. Our estimate of α also agrees with values of subgap absorption coefficients previously measured in thin films of PPV and its derivatives,[27] which ranged from 30 to 70 cm^{-1} at $\lambda = 625$ nm.

It can be seen from Fig. 7(b) that the SE threshold for $L > 1$ mm is mainly determined by α. This is an important conclusion for the quest of plastic lasers. A similar threshold condition may occur in a laser cavity where the cavity finesse Q, which determines the threshold for lasing, is limited by self-absorption: $Q \leqslant Q_{\mathrm{abs}} = 2\pi/\alpha\lambda$.[24,11] From α value measured above we estimate Q_{abs} to be about 3000; this Q value also determines the lowest attainable laser threshold and linewidth for a DOO-PPV polymer laser.[11] α in DOO-PPV films is likely to be determined by both self-absorption and scattering, and thus should vary from film to film. We conjecture that it may be possible to further decrease α and consequently lower the threshold for both ASE and lasing by improving the polymer and film qualities. In addition, other luminescent conducting polymers may have even lower optical losses, and thus may be more suitable for laser applications. Subgap α values of less than 1 cm^{-1} have been reportedly obtained in polydiacetylene films.[28] If such high optical transparency is achieved in highly luminescent conducting polymers, then Q_{abs} would be on the order of 10^5, which would, in turn, lower the SE threshold in such films by almost two orders of magnitude.

V. CONCLUSIONS

We showed that both cooperative radiation (SF) and stimulated emission (SE) contribute to the emission spectral narrowing in DOO-PPV films. SF is favored in thin films with poor optical confinement for excitation area sizes smaller than the exciton cooperation length of about 300 μm; in this case the emission pattern is isotropic. However, in DOO-PPV solutions and thin films with superior optical confinement we demonstrated directional SE. Spectral narrowing in this case was obtained by increasing either the excitation length or the excitation intensity. The excitation length and intensity dependencies of the SE intensity could be modeled using a simple one-dimensional ASE approximation. We found that for large excitation areas the SE threshold in superior DOO-PPV films is mainly determined by the optical loss coefficient at 630 nm of about 30 cm^{-1}. The SE intensity was found to rapidly increase above the threshold and quickly approach the saturation intensity of about 10^7 W/cm^2, which in turn led to gain saturation. Finally, in Table I we list the values of the gain parameters obtained using the SF and ASE models, which characterize the optical properties of DOO-PPV films.

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