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Electrically-Pumped Organic-Semiconductor Coherent Emission: A Review

F. J. Duarte

Interferometric Optics, Rochester, New York,
Department of Electrical and Computer Engineering, University of New Mexico, New
Mexico,
USA

1. Introduction

Organic lasers came into existence via the introduction of the pulsed optically-pumped liquid organic dye laser by Sorokin and Lankard (1966) and Schäfer et al. (1966). An additional momentous contribution was the discovery of the continuous wave (CW) liquid organic dye laser by Peterson et al. (1970) which opened the way for the development of narrow-linewidth tunability in the CW regime plus the eventual introduction of femtosecond lasers (see, for example, Dietel et al. (1983) and Diels, (1990)). The narrow-linewidth tunable pulsed dye laser was demonstrated by Hänsch (1972) and improved by Shoshan et al. (1977), Littman and Metcalf (1978), Duarte and Piper (1980, 1981). All these developments in practical organic tunable lasers, spanning the visible spectrum, "created a renaissance in diverse applied fields such as medicine, remote sensing, isotope separation, spectroscopy, photochemistry, and other analytical tasks" (Duarte et al. (1992)).

An early development, in the field of tunable lasers, was also the discovery of solid-state pulsed optically-pumped organic dye lasers by Soffer and McFarland (1967) and Peterson and Snavely (1968). However, it was not until the 1990s that, due to improvements in the dye-doped polymer gain media, this class of lasers would again be the focus of research attention (see, for example, Duarte (1994), Maslyukov et al. (1995), Costela et al. (2003)). An additional effort in optically-pumped tunable laser research is the work on organic semiconductor lasers based on thin-film conjugated polymers (see, for example, Holzer et al. (2002)).

All this activity has been conducted on optically-pumped organic lasers although researchers from the onset have also been interested on the direct electronic excitation of tunable organic lasers (Steyer and Schäfer, 1974; Marowsky et al., 1976). Some recent reviews mentioning efforts towards realizing coherent emission from direct electrical excitation of organic semiconductors, include Kranzelbinder and Leising (2000), Baldo et al. (2002), Samuel and Turnbull (2007), and Karnutsch (2007). Most of these reviews give ample attention to conjugated polymer gain media.

In this chapter, experimental results demonstrating coherent emission from electrically-excited pulsed dye-doped organic semiconductors, in microcavity configurations, are reviewed. The reported emission is single-transverse-mode, and given the 300 nm cavity length, also single-longitudinal mode. In the spectral domain the emission is *indistinguishable*

from broadband dye laser radiation. The radiation is generated from a tandem semiconductor structure where the emission medium are regions of coumarin 545 tetramethyl dye-doped Alq_3. An alternative description for the emission medium would be a laser-dye-doped tandem organic light emitting diode (OLED).

This work came to light in 2005 when researchers working on electrically-pumped tandem organic semiconductors reported on highly-directional coherent emission in the pulsed regime (Duarte et al., 2005). This pulsed coherent emission was characterized by a nearly diffraction limited beam and an interferometrically estimated linewidth of $\Delta\lambda \approx 10.5$ nm (Duarte et al., 2005; Duarte, 2007). In 2008 a detailed analysis of the measured emission characteristics led to the conclusion that the observed radiation was *indistinguishable* from broadband dye laser emission (Duarte, 2008). This coherent emission was generated in a sub-micrometer asymmetrical cavity comprised of a high reflector and a low reflectivity output coupler (Duarte et al., 2005; Duarte, 2007, 2008). This sub micrometer cavity was collinearly confined within an interferometric configuration which selects a single-transverse mode. The emission medium is the laser dye coumarin 545 tetramethyl.

Subsequently, using a tetramethyl dye emitting in the red, and a similar experimental arrangement, Liu et al. (2009) also reported on coherent emission in the pulsed regime. More recently, however, a paper by Samuel et al. (2009) has formulated several criticisms to the work reported by Liu et al. (2009) and interrogates their laser interpretation. Here, central aspects of Liu et al. (2009) and Samuel et al. (2009) are also reviewed and discussed in light of well-known laser, and amplified spontaneous emission (ASE), literature standards. Furthermore, the results and interpretation disclosed by Duarte et al. (2005) and Duarte (2007, 2008) are reexamined, again leading to the conclusion that the emission from the interferometric emitter is *indistinguishable* from broadband dye laser emission.

2. Coherent emission from electrically excited organic semiconductors

For completeness the salient features of the experiments discussed by Duarte et al. (2005) and Duarte (2007, 2008) are reiterated here. These experiments involve pulsed electrical excitation of organic semiconductors integrating two emitter regions in series. The active medium in each region is a coumarin 545 tetramethyl (C 545 T) dye-doped Alq_3 matrix. The structure of this class of high-brightness tandem organic semiconductors has been described in detail elsewhere (Duarte et al., 2005; Liao et al., 2004). The dye C 545 T has also been demonstrated to be a high-gain and efficient laser dye under pulsed optical excitation, by Duarte et al. (2006). Using a simple grating-mirror cavity the tuning range of this lasers is 501-574 nm. Maximum emission is observed at $\lambda \approx 555$ nm and the laser grating-narrowed linewidth is $\Delta\lambda \approx 3$ nm (Duarte et al., 2006). These results are presented in detail in Section 3.

Using the double stack electrically-excited organic light emitting diode (OLED) structure configured within an asymmetrical sub microcavity, and collinearly confined within a double interferometric structure, Duarte et al. (2005) reported on a nearly diffraction limited beam with a near-Gaussian profile and high visibility interferograms. The experimental arrangement is shown in Figure 1. The sub microcavity has a high reflectivity back mirror, that is also the cathode, and a low reflectivity output coupler, which is also the anode. This output coupler is configured by a layer of ITO and the glass interface. The external surface of the glass output coupler is antireflection coated with MgF_2 to avoid intra-glass interference. A detail description of the semiconductor structure is given in Duarte et al. (2005).
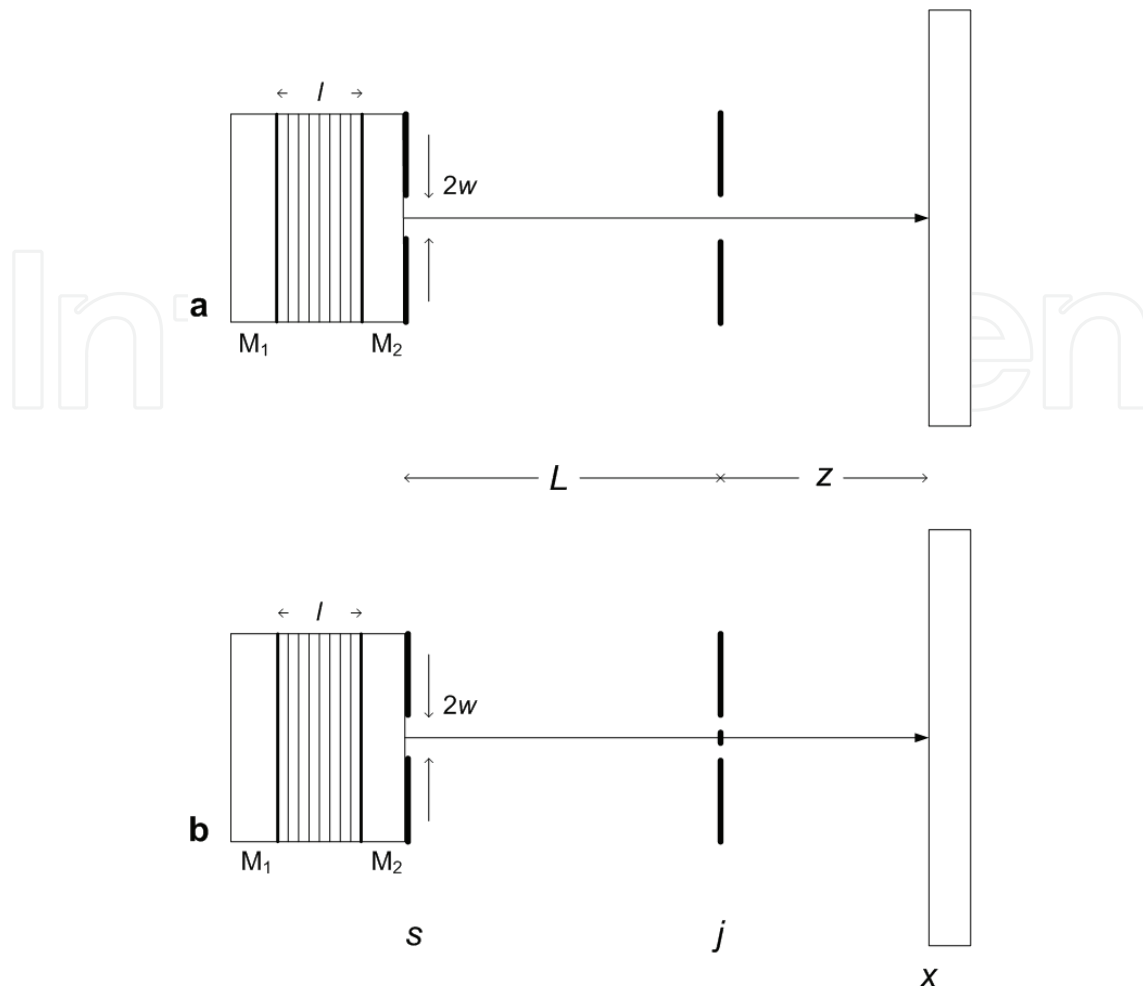
Fig. 1. (a) Electrically-pumped organic semiconductor interferometric emitter depicting the sub micrometer cavity with $l \approx 300$ nm. M_1 is a total reflector and M_2 is a low reflectivity output coupler (see text). (b) Double-slit interferometric configuration used to determine the coherence of the emission. The slits are 50 μm wide separated by 50 μm. The distance to the interferometric plane is z (from Duarte (2008)).

The overall length of this asymmetrical sub micrometer cavity is 300 nm. This interferometric emitter has been described as a *doubly interferometrically confined organic semiconductor* (DICOS) emitter where the emission medium is a laser-dye-doped Alq_3 matrix (Duarte, 2007). As described by Duarte et al. (2005) the DICOS emitter is excited with high-voltage pulses, at 100V, with ns rise times. This interferometric emitter works in the following manner: the first 150 μm aperture allows the propagation of a highly divergent, multiple-transverse-mode beam. The second $2w = 150$ μm aperture, positioned along the optical axis at $L \approx 130$ mm from the first aperture, allows propagation of a single-transverse mode exclusively. The optimum value of L is a function of wavelength and aperture dimensions (Duarte, 1993). That emission, the emission precisely along the optical axis, corresponds to a single-transverse mode, with a near-Gaussian profile (Figure 2), and exhibits a divergence near the diffraction limit as defined by the dimensions of the aperture (Duarte et al., 2005; Duarte, 2007, 2008). The digital profile of this near-Gaussian beam is shown in Figure 3.
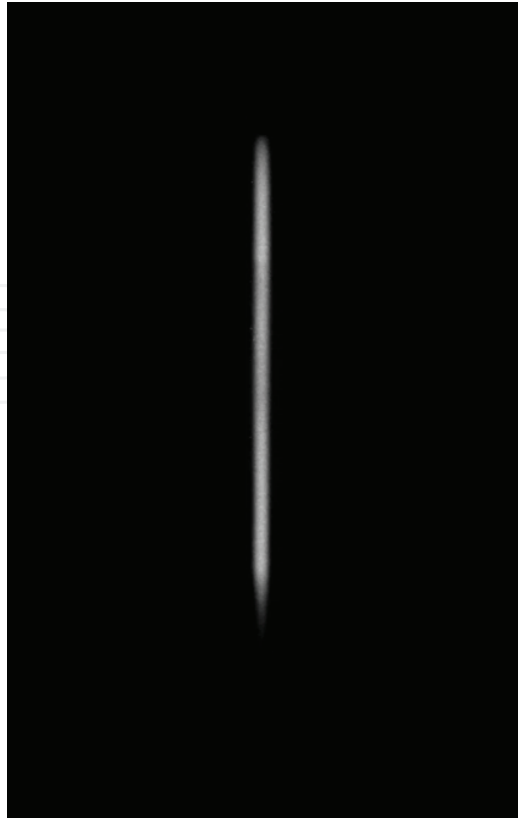
Fig. 2. Black and white silver halide photograph of the emission beam recorded at z = 340 mm. As shown in Duarte et al. (2005) the spatial profile of this single-transverse-mode emission is near-Gaussian and the beam divergence is ~ 1.1 times its diffraction limit (from Duarte et al. (2005)).
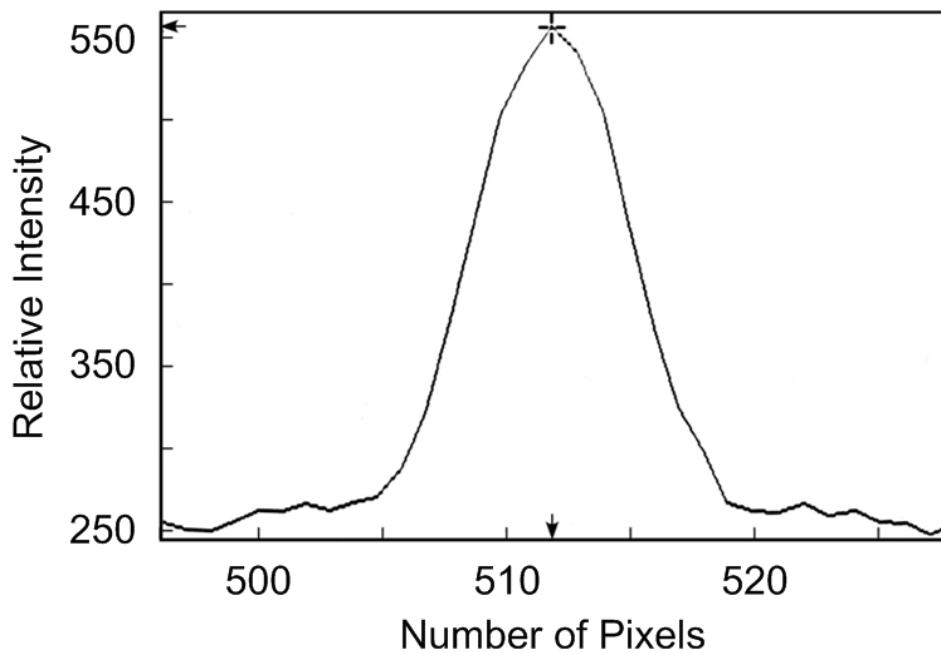

Fig. 3. Digital profile of the near-Gaussian emission beam, with a measured divergence ~ 1.1 times its diffraction limit, recorded at z = 340 mm. Each pixel is 25 μm wide (from Duarte et al. (2005)).

Considering the uncertainty in the measurement plus the uncertainty in the dimensions of the aperture a convergence towards the diffraction limit might be possible. In essence, the function of the double interferometric array is analogous to the highly discriminatory function of a multiple-prism grating configuration in narrow-linewidth laser oscillators (Duarte, 1999).

As mentioned the emission beam profile is near Gaussian and exhibits a divergence of $\Delta\theta = 2.53 \pm 0.13$ mrad which is ~ 1.1 times the diffraction limit as defined by the $2w \approx 150$ μm dimensions of the apertures (Duarte et al., 2005). The emission is also characterized by high visibility double-slit interferograms with $\mathcal{V} \approx 0.9$ (see Figure 4) which approaches the visibility regime observed from interferograms generated with the $\lambda \approx 543.30$ nm transition of a He-Ne laser with $\mathcal{V} \approx 0.95$ (see Figure 5) (Duarte, 2007). The interferometrically determined linewidth of the electrically-excited dye emission is $\Delta\lambda \approx 10.5$ nm (Duarte, 2007, 2008). Given the extremely short length of the cavity ($l \approx 300$ nm), this linewidth is consistent with single-longitudinal-mode emission since the free-spectral range is $\delta\lambda \approx 486$ nm. Pulsed output power is in the nW regime (Duarte et al., 2005) and results are summarized in Table 1. As an

$\Delta\theta$ (mrad)	$\Delta\lambda$ (nm)	$\mathcal{V}$	λ (nm)	Threshold A/cm^2
2.53[a]	~10.5[b]	0.9	~540	~ 0.8

[a] This $\Delta\theta$ corresponds to ~ 1.1 times the diffraction limit

[b] This $\Delta\lambda$ was determined using the interferometric method described in Duarte (2007, 2008).

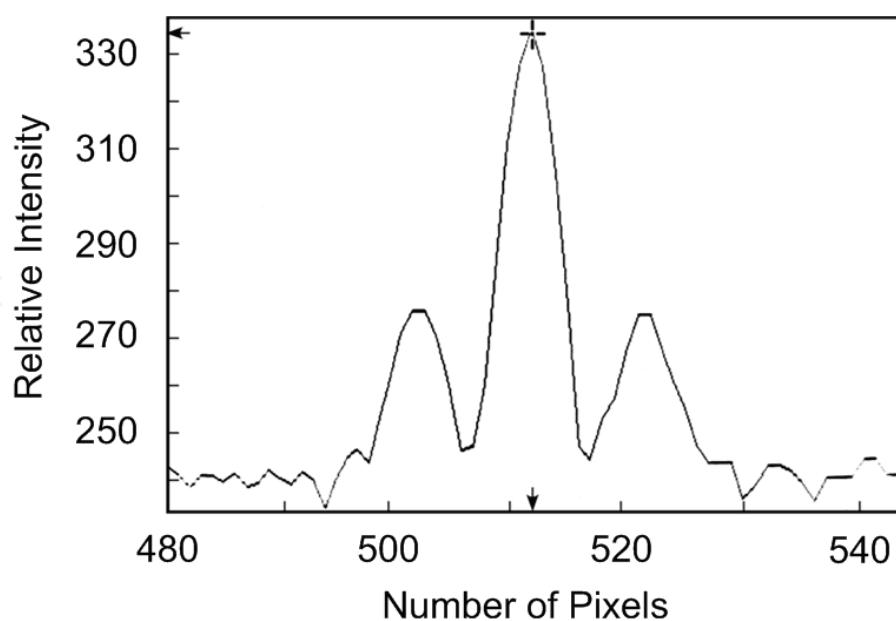Table 1. Emission parameters of the organic semiconductor interferometric emitter

Fig. 4. Double-slit interferogram of the emission from the interferometric emitter using the configuration depicted in Figure 1b. The visibility recorded at $z = 50$ mm is $\mathcal{V} \approx 0.9$ leading to an interferometrically determined linewidth of $\Delta\lambda \approx 10.5$ nm. Each pixel is 25 μm wide (from Duarte et al. (2005)).

explanatory note it should be mentioned that since both axial apertures can be physically represented as an array of a large number of sub apertures they can be considered as interferometric arrays. Indeed, interferometry of the emission is performed by replacing the second aperture by a double-slit arrangement also known as a Young-slit configuration.

Furthermore, absence of the second aperture causes the emission to be, as previously mentioned, highly divergent and multi transverse mode. The similarities of the interferograms corresponding to the electrically excited DICOS emitter and the narrow-linewidth green He-Ne laser (Figures 4 and 5) are self evident. It should be indicated that the noise in the interferogram depicted in Figure 4 is mainly detector noise given the much lower intensity levels and the fact that the digital detector was not cooled.
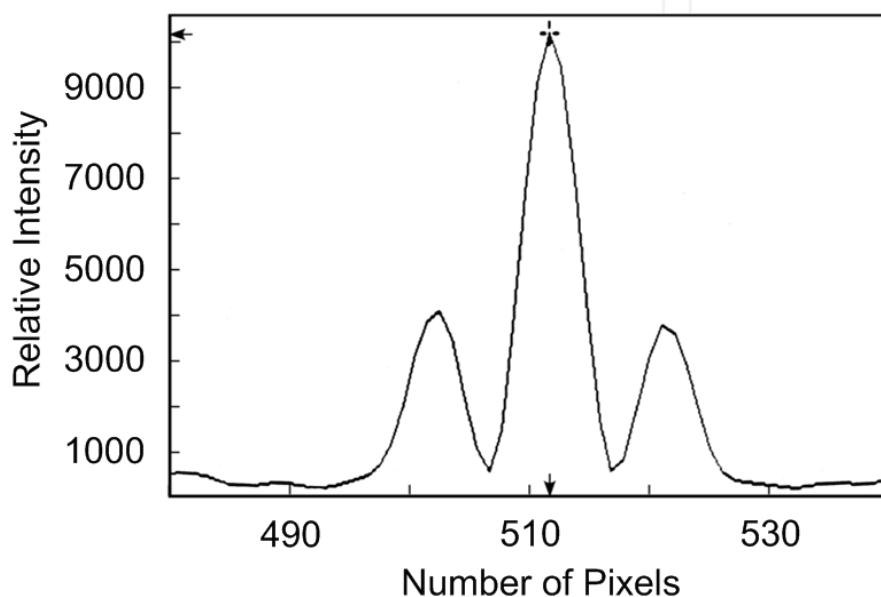

Fig. 5. Double-slit interferogram of the emission from the $\lambda \approx 543.30$ nm narrow-linewidth He-Ne laser using the interferometric configuration depicted in Figure 1b. The visibility recorded at z = 50 mm is $\mathcal{V} \approx 0.95$ while the measure laser linewidth is $\Delta\lambda \approx 0.001$ nm. Each pixel is 25 μm wide (from Duarte et al. (2005)).

3. Optically-pumped coumarin 545 tetramethyl tunable laser

As the experiments reported by Duarte et al. (2005) began it became apparent that the dye used in the tandem organic semiconductor, or tandem OLED, that is coumarin 545 tetramethyl (or C 545 T) had not been reported in the literature as a laser dye. The molecular structure of C 545 T is depicted in Figure 6. It is well known that many dyes with good to strong fluorescence characteristics might not necessarily become laser dyes. Thus, an standard laser experiment was designed to investigate the emission properties of C 545 T. If this dye was not capable of emitting coherent emission in its optically pumped version then the likelihood of observing coherent emission in the electrically-pumped regime would be infinitesimally small.

The experiment consisted in using a 3 mM solution of C 545 T in ethanol in a wedged optical cell deployed in a straight forward tunable optical cavity as depicted in Figure 7. The excitation laser is a Nitrogen laser ($\lambda \approx 337$ nm) yielding approximately 7 mJ/pulse in pulses with a duration of ~ 10 ns (FWHM).
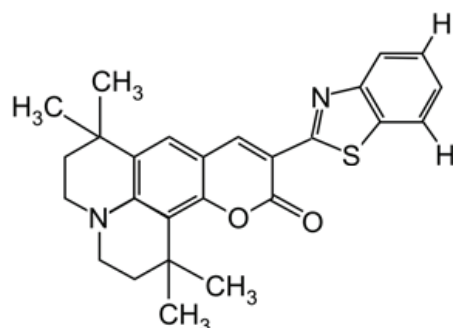
Fig. 6. Molecular structure of the laser dye coumarin 545 tetramethyl (C 545 T) (from Duarte et al., 2006).

These experiments demonstrated that C 545 T not only lased but lased extremely well under pulsed optical excitation. The measured laser efficiency was found to be ~ 14%, with a nearly diffraction limited beam divergence of $\Delta\theta \approx 1.2$ mrad., laser linewidth of $\Delta\lambda \approx 3$ nm, and an exceptional tuning range of $501 \le \lambda \le 574$ nm (see Figure 8) (Duarte et al., 2006). Thus, C 545 T adds to the excellent laser performance of the family of coumarin tetramethyl laser dyes (Chen et al., 1988; Duarte, 1989). Table 2 summarizes the performance of this optically-pumped C 545 T tunable laser.
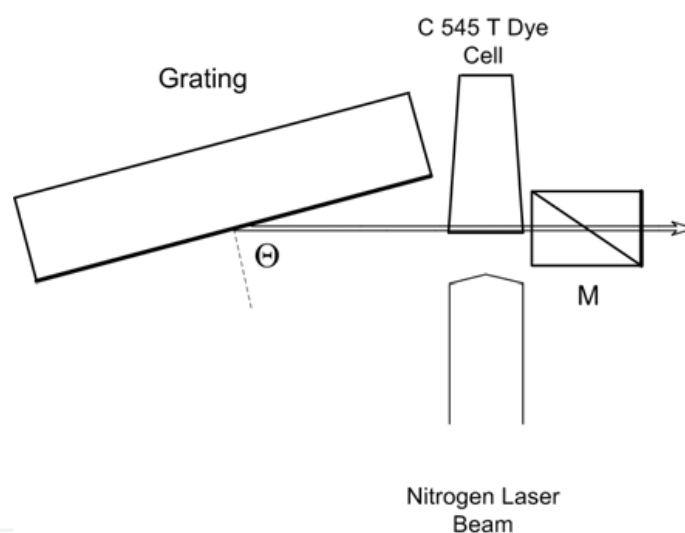

Fig. 7. Transversely-excited coumarin 545 tetramethyl dye laser. The tuning-narrowing diffraction grating has 3000 lines/mm and the output coupler-mirror is configured with a Glan-Thompson polarizer to yield laser emission polarized parallel to the plane of propagation (from Duarte et al. (2006)).

$\Delta\theta$ (mrad)	$\Delta\lambda$ (nm)	Tuning range (nm)	Efficiency %	Dye concentration mM
1.2	~3.0	$501 \le \lambda \le 574$	~14	3.0

Table 2. Performance of the grating-tuned optically-pumped C 545 T dye laser (from Duarte et al. (2006)).
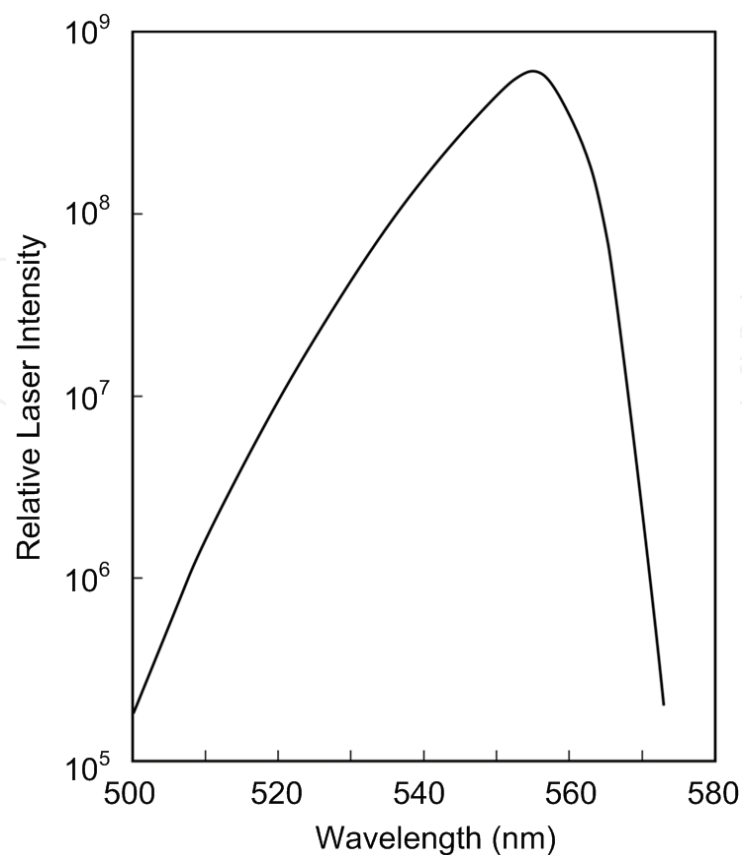
Fig. 8. Tuning curve of the transversely-excited coumarin 545 tetramethyl dye laser. The tuning range of the emission is $501 \leq \lambda \leq 574$ nm and the dynamic range of its output intensity span approximately four orders of magnitude (from Duarte et al. (2006)).

4. Microcavity emission in the red

Using an experimental configuration partially similar to that disclosed by Duarte et al. (2005) and Duarte (2007, 2008) (without the second aperture), Liu et al. (2009) reported on laser emission at 621.7 nm using a red emitting tetramethyl dye-doped active medium. A summary of this report includes a linewidth of $\Delta\lambda \approx 1.95$ nm, a beam divergence of $\Delta\theta \approx 32$ mrad, interferometric visibility of $\mathcal{V} \approx 0.89$, and a current threshold of 0.86 A/cm.[2] However, in a recent paper Samuel et al. (2009) interrogate various aspects of Liu et al. (2009), including:

1. The linewidth reduction from 2.62 nm, below threshold, to 1.95 nm, above threshold, is deemed as insufficient evidence of lasing. An analogous comment is made in reference to beam divergence (Samuel et al., 2009).
2. The threshold current density of 0.86 A/cm[2] is said to be "five orders of magnitude smaller" than expected (Samuel et al., 2009).
   Thus, the output emission reported in Liu et al. (2009) is not classified by Samuel et al. (2009) as corresponding to laser emission. In a more general context Samuel et al. (2009) highlight the importance of polarization in organic laser emission and formulate further assertions including:
3. "Interference effects can be observed perfectly well using a lamp and a pair of slits... The observation of interference phenomena is intriguing, but the source is small and

therefore capable of giving high-visibility fringes when illuminating double slits" (Samuel et al., 2009).

4.   "The typical linewidth of ASE in an organic semiconductor is 10 nm" (Samuel et al., 2009). Here, the comments and assertions of Samuel et al. (2009) are examined in light of the published dye laser literature in Sections 5, and 7-9.

5. Microcavity lasers and thresholds

Experiments published on optically-pumped liquid dye lasers with a cavity length $l \leq \lambda$ indicate that a near "zero-threshold-laser" emission is observed for $l \approx \lambda / 2$ (De Martini et al., 1988). These results were obtained with a multiple-transverse-mode emission beam (De Martini et al., 1988). Albeit the beam waist, on focus at the gain region, of the excitation beam is not given by these authors, it can be shown that even with an emission beam waist of only a few micrometers there would be an enormous number of transverse modes due to the $l \approx \lambda / 2$ length of the cavity. The relevant fact here is that De Martini et al. (1988) did observe a near "zero-threshold-laser" emission under multiple-transverse-mode conditions. These experiments provide very persuasive evidence in support of threshold behavior at very low excitation densities. In other words, the experiments of De Martini et al. (1988) strongly suggest that with a suitable gain medium, and microcavity configurations, high threshold energy densities are not required. Subsequently, with a suitable gain medium, and microcavity, current density thresholds in the 10-100 kA/cm² range, as mentioned by Samuel et al. (2009), should not be necessary. Therefore, low threshold behavior at 0.8–0.9 A/cm² (Duarte et al., 2005; Duarte, 2007, 2008; Liu et al., 2009) is consistent with what would be expected in a sub micrometer cavity where the conditions $l < \lambda$ do apply. It should also be mentioned that in our own experiments rapid destruction of the laser dye-doped semiconductor medium was observed at peak excitation voltages, in the nanosecond regime, approaching 10 kV corresponding to current densities of only ~ 190 A/cm² (Duarte, 2008). The emission beam profile under such extreme excitation conditions is shown in Figure 9.
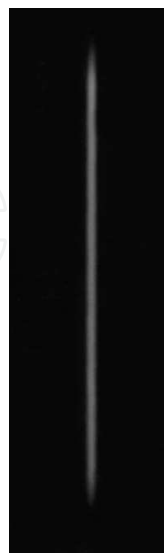

Fig. 9. Black and white silver halide photograph of the near-Gaussian emission beam, from the interferometric emitter (DICOS), recorder under nanosecond pulsed excitation at ~ 10 kV per pulse. The corresponding excitation current density is ~ 190 A/cm.² (from Duarte (2008)).

6. Beam divergence and transverse mode structure

The ideal *diffraction limited* beam divergence, derived from the uncertainty principle $\Delta x \Delta p \approx h$ (Dirac, 1978), is given by Duarte (2003)

$$\Delta\theta \approx (\lambda / \pi w) \tag{1}$$

where λ is the emission wavelength and w the emission beam waist. However, the beam divergence from a cavity can be augmented by geometrical factors included in the expression (Duarte, 1990a)

$$\Delta\theta \approx (\lambda / \pi w) \left(1 + (L_R / B)^2 + (L_R A / B)^2 \right)^{1/2} \tag{2}$$

where

$$L_R = (\pi w^2 / \lambda)$$

is the Rayleigh length and A and B are propagation terms from propagation matrix theory (Duarte, 1989, 1990a). In well-designed narrow-linewidth laser oscillator cavities the beam divergence often approaches the diffraction limit as the term in parenthesis approaches unity (Duarte, 1990a, 1999). For a complete matrix treatment of tunable laser resonators the reader should refer to Duarte (1989, 1992, 2003).

For a simple mirror-mirror resonator, in the absence of intracavity beam expansion $A \approx 1$ and B becomes the intra cavity length l (see Figure 1), so that $B = l$, and

$$\Delta\theta \approx (\lambda / \pi w) \left(1 + 2(L_R / l)^2 \right)^{1/2} \tag{3}$$

For a microcavity the condition $l \leq \lambda$ applies, and $L_R >> l$, so that

$$\Delta\theta >> (\lambda / \pi w) \tag{4}$$

Thus, large beam divergences are inherent to resonators where the cavity length is in the sub micrometer, or nanometer, regime as in the case of what is understood for sub micrometer cavities where the condition $l \leq \lambda$ applies. Thus the use of a secondary aperture along the propagation axis, as depicted in Figure 1, is necessary if near single-transverse-mode emission is desired.

A more accurate description of the diffraction limited divergence as given in (Duarte, 2008)

$$\Delta\theta \approx (\lambda \pm \Delta\lambda) / (\pi w) \tag{5}$$

where $\Delta\lambda$ is the usual linewidth of emission. In this regard, a cumulative spatial detector (such as a silver-halide photographic plate) registers

$$\Delta\theta \approx (\lambda + \Delta\lambda) / (\pi w) \tag{6}$$

which illustrates to a first approximation that the narrowing in the spectral width, as the emission transitions from ASE to lasing, leads to a decrease in the beam divergence.

The conditions outlined in Equation (4) lead to an emission characterized by a very large number of transverse modes as can be quantified via the interferometric equation (Duarte, 1993, 2007, 2008)

$$|\langle x|s\rangle|^2 = \sum_{j=1}^{N}\Psi(r_j)^2 + 2\sum_{j=1}^{N}\Psi(r_j)\left(\sum_{m=j+1}^{N}\Psi(r_m)\cos(\Omega_m - \Omega_j)\right) \tag{7}$$

where the phase term in parenthesis relates directly to the exact geometry of the cavity and the wavelength of emission (Duarte, 2003). Moreover, because the cavity is so short the free spectral range, given by

$$\delta\lambda \approx \lambda^2 / (2l) \tag{8}$$

is very wide ($\delta\lambda \approx 486$ nm). Thus for emission linewidths in the 10 nm range (as in Duarte et al. (2005) and Duarte (2007, 2008)), only single-longitudinal-mode emission for each transverse mode is allowed.

Therefore, for $l \approx 300$ nm, and $2w \approx 150\,\mu$m, the experimenter using a conventional configuration is confronted with large beam divergences and a multitude of transverse modes. One way to overcome this compounded problem is to position, at a distance determined by Equation (7), a second slit to filter out all the unwanted modes and allow only a single-transverse-mode that, as discussed previously, under appropriate linewidth conditions, would only allow a single-longitudinal mode. This concept led to the doubly interferometrically confined organic semiconductor (DICOS) emitter described in (Duarte et al., 2005; Duarte, 2007, 2008) and depicted in Figure 1.

Equation (7) is also used to generate a series of numerically based interferograms, using the emission wavelength and interferometric parameters applicable to the interferometric emitter, which are used in a graphical technique to estimate the approximate linewidth of the emission (see Duarte (2007, 2008)). This interferometrically determined linewidth yields a conservative upper limit for the emission linewidth, in this case $\Delta\lambda \approx 10.5$ nm (Duarte, 2007, 2008).

7. Visibility of double-slit interferograms

It is well-known that "interference effects can be observed perfectly well using a lamp and a pair of slits" (Samuel et al., 2009). Indeed, in their classic experiment Thompson and Wolf (1957) observed beautiful fringes with $\mathcal{V} \approx 0.593$ from a simple mercury lamp. Nevertheless, the literature indicates that interferograms with high visibility, $\mathcal{V} \geq 0.9$, are coherently speaking more special. In a directly relevant experiment while using a similar geometry as (Duarte et al., 2005; Duarte, 2007, 2008), and a small organic semiconductor source, Saxena et al. (2006) have reported on visibilities of $\mathcal{V} \approx 0.4$ which is related to a measured linewidth of $\Delta\lambda \approx 100$ nm. Experiments on the measurement of interferometric visibility for dye ASE sources give a visibility of $\mathcal{V} \approx 0.65$ which is related to a measured linewidth of $\Delta\lambda \approx 17$ nm (Dharmadhikari et al., 2005).

The use of double-slit interference techniques, to characterize the coherence of laser emission, is a well-known and accepted practice documented in the laser literature (Nelson and Collins, 1961; Shimkaveg et al., 1992; Trebes et al. 1992; Ditmire et al., 1996; Lucianetti et

al., 2004). Indeed, double-slit interferometric measurement have been used to characterize laser emission since the dawn of the laser age (Nelson and Collins, 1961). In this regard, laser emission is associated with visibilities in the approximate $0.85 \leq \mathcal{V} \leq 1.0$ range (Trebes et al., 1992; Lucianetti et al., 2004). In our own experiments we have measured a visibility of $\mathcal{V} \approx 0.90$ using the configuration depicted in Figure 1b and $\mathcal{V} \approx 0.95$ for the equivalent interferometric configuration while using illumination from the narrow-linewidth $3s_2$–$2p_{10}$ transition ($\Delta\lambda \approx 0.001$ nm) of the He-Ne laser at $\lambda \approx 543.3$ nm (Duarte et al., 2005; Duarte, 2007; 2008).

Thus the existing literature documents that the use of double-slit interferograms is a well accepted practice in determining the coherence of laser sources since 1961. It is further well established that for typical ASE and non laser emission the measured visibilities are below $\mathcal{V} \approx 0.65$. The published literature also indicates that emission with visibility in the range of $0.85 \leq \mathcal{V} \leq 1.0$ is highly coherent and therefore associated with laser emission. Besides citing the refereed literature, and in addition to the comparison with the coherence from the green He-Ne laser, we now refer to a direct experiment that registered the interferograms from the optically-pumped high-power C 545 T dye laser and the electrically-pumped C 545 T dye-doped interferometric emitter. The interferograms were recorded directly on black and white silver-halide film thus leaving a permanent photographic record of the experiment (Duarte, et al., 2005; Duarte, 2008).
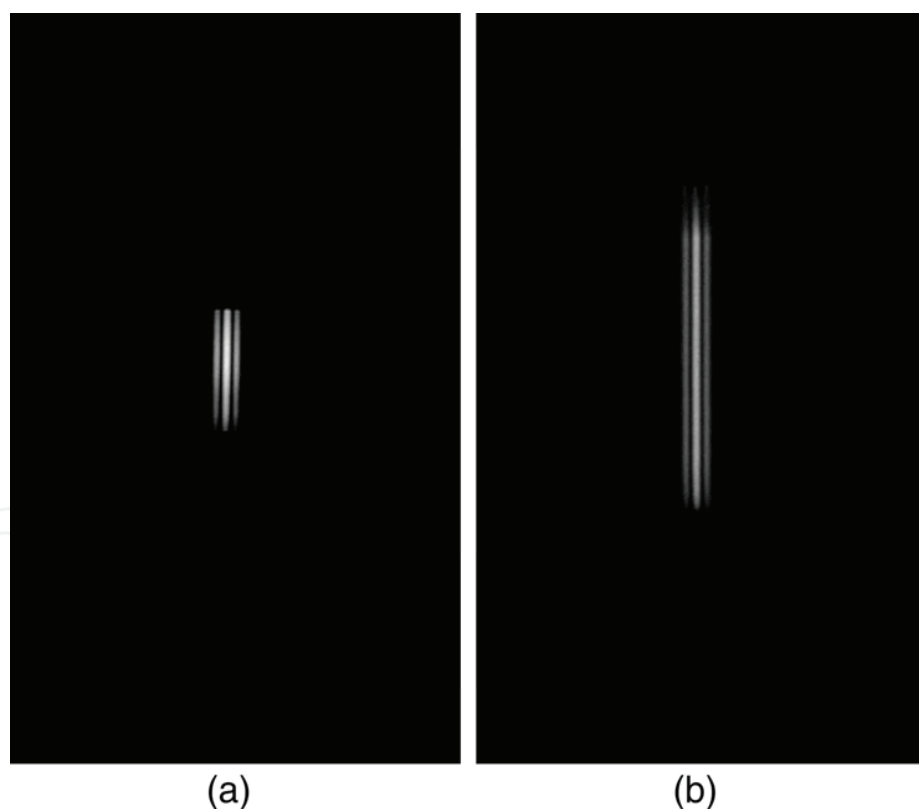

Fig. 10. Back and white silver halide photographs of double-slit interferograms produced by (a) laser emission from a grating-narrowed C 545 T dye laser (Duarte et al., 2006) and (b) from the C 545 T dye-doped electrically-excited organic semiconductor interferometric emitter as depicted in Figure 1b. In both cases the slits are 50 μm wide, separated by 50 μm, and $z = 175$ mm (from Duarte et al. (2005)).

Figure 10 shows the interferograms recorded under identical interferometric conditions, for the optically-pumped C 545 T dye laser (with $\Delta\lambda \approx 3$ nm) (Duarte et al., 2006), and the electrically-excited interferometric emitter (depicted in Figure 1b). Although these interferograms, at first glance, appear to be quite similar, examination under magnification reveals slightly broader characteristics from the interferometric emitter that are quite consistent with the interferometric estimate (Duarte, 2007, 2008) of $\Delta\lambda \approx 10.5$ nm.

8. Emission bandwidth of ASE versus laser emission linewidth

The presence of ASE, in narrow-linewidth dye lasers, is a physics and engineering challenge that has been a subject of sustained interest among researchers for quite a while (Duarte, 1990, 2003). In particular, optimized oscillator cavities yielding tunable narrow-linewidth laser emission with extremely low levels of ASE have been successfully developed and optimized (Duarte and Piper, 1980; Duarte et al., 1990). In this effort it has been learned that ASE in dye lasers can be as wide as 50-60 nm (Dujardin and Flamant, 1978; Duarte and Piper, 1980; Bor, 1981; McKee et al., 1982) and can be successfully suppressed, in optimized multiple-prism grating cavities, to levels as low as

$$(\rho_{ASE} / \rho_{laser}) \approx 10^{-9}$$

for highly-coherent, highly-polarized, laser emission with $\Delta\nu \approx 360$ MHz at 590 nm (or $\Delta\lambda \approx 0.00042$ nm) (Duarte et al., 1990). Here it should be noted that the highly polarized emission (parallel to the plane of propagation) is almost entirely a by-product of the cavity architecture (Duarte et al., 1990; Duarte, 1990a).

On the other hand, dye lasers are also capable of emitting high-power emission in a broadband mode while using basic *mirror-mirror* resonators. Iconic examples of such lasers are the dye laser reported by Schäfer et al. (1966), with a bandwidth of $\Delta\lambda \approx 10$ nm, and the dye lasers reported by Spaeth and Bortfeld (1966) with emission bandwidths in the $4.5 \leq \Delta\lambda \leq 7$ nm range.

In summary, pulsed laser dye ASE can be as wide as 50-60 nm (Dujardin and Flamant, 1978; Duarte and Piper, 1980; Bor, 1981; McKee et al., 1982), and high-power dye lasers can be designed to either deliver broadband laser emission in the approximate $4.5 \leq \Delta\lambda \leq 10$ nm range (Schäfer et al., 1966; Spaeth and Bortfeld, 1966; Baltakov et al., 1973), or highly coherent emission with laser linewidths as narrow as $\Delta\lambda \approx 0.0004$ nm at 590 nm (Duarte, 1999). The characteristics of the emission depend on various parameters including the excitation conditions, laser dye concentration, and very importantly, the resonator architecture. Again, dye lasers can emit in the ASE regime (pre-laser emission), the broadband laser regime, and the narrow-linewidth laser regime.

9. Polarization in organic dye lasers

Shäfer (1990) and Duarte (1990b) provide detailed discussions on the polarization characteristics of laser and ASE emission in dye lasers. It is beautifully illustrated, for instance, that the intrinsic polarization orientation of the emission from a particular dye can be controlled by choosing the orientation of the polarization of the excitation laser relative to the propagation plane of the dye emitter (Duarte, 1990b). In the case of the copper-laser-

pumped rhodamine 590 dye laser these polarization alternatives enable the designer to engineer a broadband dye laser with either an emission polarization perpendicular to the plane of propagation or nearly undefined (that is, nearly unpolarized). Furthermore, the overall final polarization for narrow-linewidth laser emission depends critically on various other factors. Thus, for laser-pumped dye lasers the final outcome is a function of the orientation of the polarization of the pump laser, the polarization preference of the grating, the orientation of the windows of the active region, the degree of polarization selection of the beam expander, and the presence or absence of an intracavity polarizer (Duarte, 1990b). Therefore, one avenue to optimize the overall conversion efficiency of a laser oscillator consists in carefully matching the polarization preference of all the above mentioned parameters to yield efficient, highly polarized, narrow-linewidth laser emission (Duarte and Piper, 1984). One recent example of such optimization is the narrow-linewidth solid-state multiple-prism grating laser oscillator shown in Figure 11. The rhodamine 6G dye doped polymer matrix is configured to favor polarization parallel to the plane of propagation, this polarization is then reinforced by the multiple-prism beam expander, the 3300 line/mm diffraction grating, and the Glan-Thompson polarizer output coupler (Duarte, 1999). This dispersive oscillator yields single-transverse-mode, single-longitudinal-mode, highly-polarized laser emission (parallel to the plane of propagation) with a linewidth of $\Delta v \approx 350$ MHz (see Figure 12) and a pulse duration of $\Delta t \approx 3$ ns which means that the emission is nearly limited by Heisenberg's uncertainty principle.
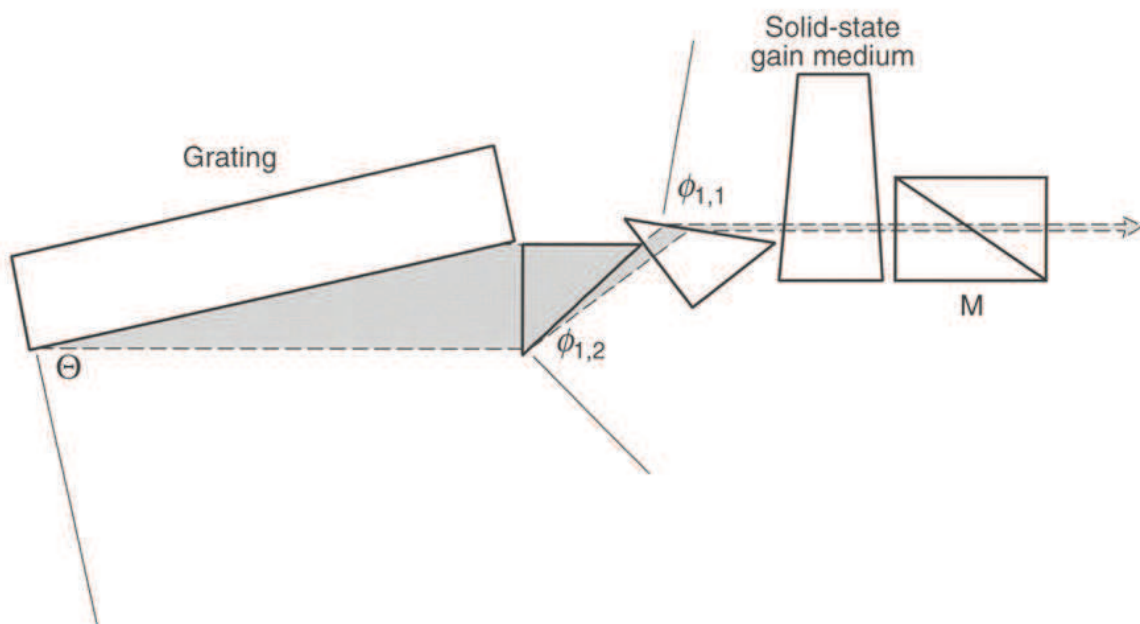

Fig. 11. Multiple-prism grating solid-state dye laser oscillator yielding single-transverse-mode and single-longitudinal-mode laser emission at a linewidth of $\Delta v \approx 350$ MHz ($\Delta \lambda \approx 0.0004$ nm at 590 nm) at extremely low levels of ASE. The temporal pulse (~ 3 ns at FWHM) is nearly Gaussian and its tuning range is $550 \leq \lambda \leq 603$ nm. Due to the polarization preference of the grating, the multiple-prism beam expander, and the Glan-Thompson polarizer output coupler, the emission is highly polarized parallel to the plane of propagation. The gain medium is rhodamine 6 G dye-doped PMMA at a concentration of 0. 5 mM (from Duarte (1999)).
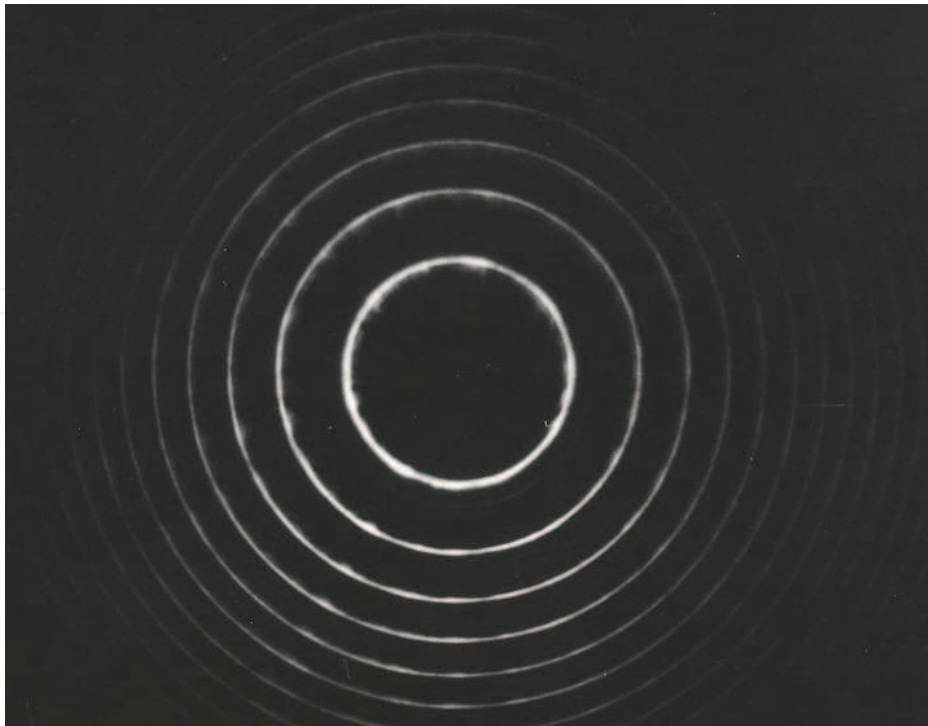
Fig. 12. Fabry-Perot interferogram of the single-longitudinal-mode emission, at
$\Delta v \approx 350$ MHz, from the optimized solid-state multiple-prism grating laser oscillator (from Duarte (1999)).

The ASE level of this highly polarized single-longitudinal-mode emission is quoted as $\sim 10^{-6}$. The polarization discussion in this section is aimed at reminding the readership that, in organic dye lasers, polarization of the emission depends principally on the cavity architecture.

10. Additional comments on broadband laser emission

One of the characteristics of dye gain media is its high gain. The original reason to introduce trapezoidal dye optical cells (Duarte, 1980) was to successfully *avoid broadband lasing* due to the resonator cavity effect provided by the uncoated windows. With the trapezoidal geometry the excited dye region emits only ASE along the optical axis. This ASE can then be guided through the intracavity dispersive optics so that, following proper alignment of the cavity, low noise tunable narrow-linewidth emission can be achieved (Duarte, 1980, 1984, 1999).

A subtle point emerges here that is worth further attention. When broadband lasing occurs, due to the low reflectivity of parallel uncoated windows, and despite the absence of external mirrors, it is because a resonator cavity has been configured by the uncoated windows. Indeed, it is improper to label this broadband lasing, contained in a directional laser beam, as ASE. Certainly, the same effect occurs in solid-state dye lasers leading to the use of dye-doped polymer matrices configured in trapezoidal geometries (Duarte, 1994, 1999). As a footnote it should be mentioned that once broadband lasing occurs, due to the low-reflectivity effect mentioned here, it is not possible to bring the emission under control with the external optics shown in Figure 11. Hence, it is imperative to suppress broadband lasing prior to achieving controllable low noise narrow-linewidth emission.

11. Additional comments on the microcavity emission in the red

As already indicated by Samuel et al. (2009), Liu et al. (2009) adopted an experimental approach similar to that of Duarte et al. (2005). In particular, they use double slit interference to determine that the emission interferograms yield a visibility of $\mathcal{V} \approx 0.51$ prior to threshold and a $\mathcal{V} \approx 0.89$ post threshold (Liu et al., 2009). This post-threshold visibility is close to the value reported by Duarte et al. (2005) and Duarte (2007, 2009) of $\mathcal{V} \approx 0.9$. The interferogram published by Liu et al. (2009) is perfectly symmetrical and devoid of noise. If this represents a series of averaged interferograms it is not mentioned by the authors (Liu et al., 2009), however, in our experience a perfectly-symmetrical noiseless interferogram would be an extremely rare occurrence (see Figures 4 and 5).

The magnitude of the change in interferometric visibility reported in (Liu et al., 2009), from $\mathcal{V} \approx 0.51$ to $\mathcal{V} \approx 0.89$, is not consistent with the relatively small transition in linewidth, from 2.36 nm to 1.95 nm. From an alternative perspective a bandwidth of 2.26 nm is unusually narrow to be associated with dye ASE as discussed in the previous section. Moreover, the reported pre threshold visibility of $\mathcal{V} \approx 0.51$ is too low to be associated to a linewidth of 2.26 nm.

12. Discussion

Recently, Newton's ring interferometry has been used on standard C 545 T dye-doped OLED devices yielding measured linewidths in the $40 \leq \Delta\lambda \leq 110$ nm range with the authors attributing the narrower linewidth ($\Delta\lambda \approx 40$ nm) to "microcavity effects" (Tsia et al., 2010). These results are consistent with those of Saxena et al. (2006) who reported $\Delta\lambda \approx 100$ nm and $\mathcal{V} \approx 0.4$ for their organic semiconductor device. These completely independent results illustrate that the high degree of coherence reported in Duarte et al. (2005) and Duarte (2007, 2008), is not observed in the standard OLED devices of Saxena et al. (2006) or Tsia et al. (2010). For emission originating from a laser dye medium, such as C 545 T, it is experimentally justified to expect, according to the literature, a wide spectral coverage for ASE (50-60 nm) (Dujardin and Flamant, 1978; Duarte and Piper, 1980; Bor, 1981; McKee et al., 1982) and relatively broad laser linewidths, in the approximate $4.5 \leq \Delta\lambda \leq 10$ nm range, for simple mirror-mirror cavities (Schäfer et al., 1966; Spaeth and Bortfeld, 1966; Baltakov et al., 1973). By contrast, Samuel et al. (2009) mention that "typical linewidth of ASE in an organic semiconductor is 10 nm." Although this statement might be relevant for conjugated polymer gain media it does not apply to laser-dye based gain media (Dujardin and Flamant, 1978; Duarte and Piper, 1980; Bor, 1981; McKee et al., 1982).

The visibility of the interferograms $\mathcal{V} \approx 0.90$ (Duarte et al., 2005; Duarte, 2007, 2008) is considerably higher than the visibilities associated with standard OLED devices (Saxena et al., 2006; Tsia et al., 2010), higher than the visibility associated with ASE (Dharmadhikari et al., 2005), and consistent with the interferometric visibilities of laser emission (Trebes et al., 1992; Lucianetti et al., 2004). Further, the interferometrically determined linewidth $\Delta\lambda \approx 10.5$ nm is consistent with the linewidth reported for broadband dye laser emission in the iconic paper of Schäfer et al. (1966). As a result of the double interferometric confinement (Figure 1b) the emission radiation corresponds to a single-transverse-mode (Figures 2 and 3) and a single-longitudinal-mode as discussed in Section 6. As suggested in (Duarte, 2007) the likely origin for the observed spectral coherence is the resonance established in the asymmetrical sub microcavity, or nanocavity. In this regard, under excitation with a fast

rise time pulse (Duarte et al., 2005; Duarte, 2007) it would only take a tiny fraction of a nanosecond for the emission to execute an enormous number of intracavity return passes since $l \approx 300$ nm. Previous studies on optically-pumped solid-state narrow-linewidth dye laser oscillators, using asymmetrical cavity configurations, established that only a few intracavity return passes are necessary to reach laser threshold (Duarte, 2003).

The lack of access, via mirror controls, associated with the class of sub microcavity configuration used in these experiments, does deny the opportunity to observe the emission as a function of cavity alignment. As previously indicated (Duarte, 2008) this might require the construction of a series of devices with various output coupler reflectivities in addition to changing, by design, the angle of incidence, relative to the optical axis, at the output coupler mirror. Since mirror dependence of the emission was not demonstrated an explicit claim of a conventional laser device, or "an electrically-pumped dye laser," was not made in Duarte et al. (2005) or Duarte (2007, 2008). Nevertheless, the experimental evidence clearly shows that the coherent emission reported in these papers (Duarte et al., 2005; Duarte, 2007, 2008) is, spatially and spectrally, indistinguishable from broadband dye laser emission.

13. Conclusion

In this chapter experimental results demonstrating spatially coherent, and interferometrically coherent, emission from pulsed electrically-pumped coumarin 545 tetramethyl laser-dye-doped organic semiconductors have been reviewed. This review also takes into account interpretational issues brought forward in recent publications. In particular, this review re-examines concepts and parameters relevant to amplified spontaneous emission, linewidth, and polarization in tunable organic dye lasers. This has been done by revisiting published results on dye amplified spontaneous emission, broadband dye lasers, and narrow-linewidth tunable laser oscillators using laser dye gain media. This has been performed using seminal and historically significant references from the peer-reviewed literature. This exercise leads us to reaffirm the previous interpretation of Duarte et al. (2005) and Duarte (2007, 2008) that the emission observed from the electrically-excited doubly interferometrically confined organic semiconductor emitter is, coherently speaking, *indistinguishable from broadband dye laser emission*.

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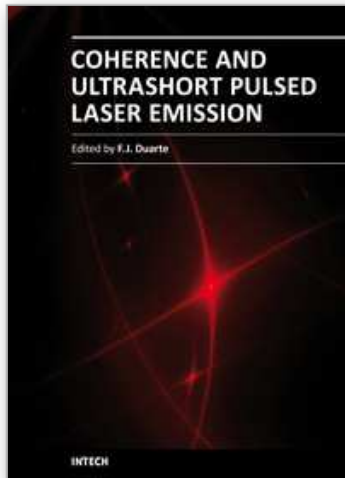
Coherence and Ultrashort Pulse Laser Emission

Edited by Dr. F. J. Duarte

In this volume, recent contributions on coherence provide a useful perspective on the diversity of various coherent sources of emission and coherent related phenomena of current interest. These papers provide a preamble for a larger collection of contributions on ultrashort pulse laser generation and ultrashort pulse laser phenomena. Papers on ultrashort pulse phenomena include works on few cycle pulses, high-power generation, propagation in various media, to various applications of current interest. Undoubtedly, Coherence and Ultrashort Pulse Emission offers a rich and practical perspective on this rapidly evolving field.

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