Chapter 1

Introduction

F J Duarte

1.1 Introduction

Organic dye lasers were discovered by Sorokin and Lankard (1966) and Schäfer *et al* (1966). Both these reports refer to pulsed laser-pumped liquid gain media, comprised of diluted organic dye molecules in a organic solvent, situated within a laser resonator comprised of two mirrors.

The next grand entrance into the laser field, via the organic laser, was performed by the *tunable* organic dye laser (Spaeth and Bortfeld 1966, Soffer and McFarland 1967). A few years later, followed the very first *tunable narrow-linewidth* laser introduced by Hänsch (1972). These groundbreaking disclosures set in motion an enormous wave of discoveries with gigantic innovative consequences to analytical chemistry, astronomy, medicine, nuclear industry, physics, spectroscopy, and science in general. A wave of innovation that remains vastly uncelebrated with most of its innovators only tenuously and vaguely recognized.

It is the tunability, throughout the visible spectrum, and the exquisite specificity in wavelength selectivity that made the tunable organic dye laser such a worldwide success in the scientific laboratories of the 1970s-to-1990s epoch. In addition to this tunable coherent ability, the organic dye laser in the liquid state is uniquely apt to remove excess heat from an inherently high-gain molecular active medium that has led to the demonstration of single-pulse energies in the 400–800 J (Baltakov *et al* 1974, Tang *et al* 1987) range and average powers exceeding 25 kW (Bass *et al* 1992). An additional landmark was the introduction of the continuous wave (CW) organic dye laser (Peterson *et al* 1970) that became the developmental workhorse for the femtosecond dye laser (Fork *et al* 1981, 1987, Dietel *et al* 1983) and a background model for ultrafast lasers in general.

The early revolution of tunable organic lasers, and laser dyes, is well covered in various books including *Dye Lasers* (Schäfer 1990), *Laser Dyes* (Maeda 1984), *Dye Laser Principles* (Duarte and Hillman 1990), *High-Power Dye Lasers* (Duarte 1991), and *Selected Papers on Dye Lasers* (Duarte 1992).

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The whole gamut of organic lasers now is extended to include solid-state tunable organic dye lasers and semiconductor organic lasers. This latest generation of organic lasers has opened the horizon to the realm of miniaturized devices with its own vast array of applications.

Organic Lasers and Organic Photonics is the very first book to include all these wonderful sources of tunable coherent radiation integrated in a single cohesive, and unique, volume. Furthermore, a pragmatic description of organic photonics, including a plethora of exciting applications of organic lasers, is given in addition to stand-alone chapters on optogenetics, organic laser medicine, and quantum communications.

1.2 Laser linewidth

A laser is differentiated, recognized, and desired, by its ability to emit coherent radiation: that is, spatial coherence and spectral coherence. Spatial coherence is what gives the laser its ability to direct its output energy towards a single confined, minute, area in space equal to the cross section of its emission beam. The first beam divergence from an organic dye laser was quoted by Sorokin and Lankard (1966) and it was 3 mrad (at half angle) at a wavelength of $\lambda \approx 755$ nm.

Spectral coherence is what gives the laser its ability to emit in extremely narrow segments of the electromagnetic spectrum or in extremely pure colors. A measure of how narrow is that segment of the electromagnetic spectrum, or how pure is the color of emission, is the *laser linewidth*. In this section attention is given to what is laser linewidth and what constitutes narrow-linewidth emission in organic lasers. One observation worth mentioning at this stage is that, in regard to the quality of emission, the concept of coherence is more restrictive than the concept of laser. For instance, a device described as a *laser* can have multiple transverse modes and each of those transverse modes can include a multitude of longitudinal modes. Thus a basic laser can be a mediocre source of coherent radiation. On the other hand, a device described as a *coherent source* or *coherent emitter* implies coherence both in the spatial and the spectral domain and thus it can be assumed to emit a single-longitudinal mode in a single-transverse mode.

1.2.1 Laser linewidth in organic dye lasers

As previously mentioned, the first unambiguously measured emission linewidths in organic dye lasers were published by Soffer and McFarland (1967): $\Delta\lambda \approx 6$ nm, at a wavelength of $\lambda \approx 570$ nm for a mirror-mirror cavity, and $\Delta\lambda \approx 0.06$ nm at a wavelength of $\lambda \approx 565$ nm for a mirror-grating cavity. A linewidth of $\Delta\lambda \approx 6$ nm would be classified as broadband laser emission while, on the other hand, a linewidth of $\Delta\lambda \approx 0.06$ nm would begin to *approach the realm of narrow-linewidth* laser emission. A brief literature survey indicates that in high-peak-power pulsed liquid organic dye lasers the laser linewidth, in the absence of intracavity etalons, can be in the $0.000\,45 \leqslant \Delta\lambda \leqslant 6$ nm range (Soffer and McFarland 1967, Duarte and Piper 1984).

Usually in optimized narrow-linewidth laser oscillators, the narrowness of the laser linewidth $\Delta\lambda$, or $\Delta\nu$ in Hz units, is limited by the pulse duration Δt of the laser emission via a consequence of Heisenberg's uncertainty principle or $\Delta\nu\Delta t \approx 1$.

1.2.2 Narrow-linewidth landmarks in high-power pulsed organic dye lasers

Here, landmark contributions and discoveries pertinent to the development of high-power pulsed tunable narrow-linewidth organic dye lasers are listed. These discoveries albeit originally introduced in the field of organic lasers are found extensively applied throughout laser technology in general.

1967: Intracavity diffraction grating tuning is introduced while also demonstrating dispersion induced linewidth narrowing (Soffer and McFarland 1967).

1971: The distributed feedback (DFB) organic dye laser is discovered (Kogelnick and Shank 1971).

1972: Introduction of the first narrow-linewidth tunable organic dye laser also demonstrating grating dispersion multiplication via intracavity telescopic beam expansion (Hänsch 1972).

1977–78: Discovery of grazing-incidence grating cavities (Shohan *et al* 1977, Littman and Metcalf 1978, Saikan 1978).

1978–80: Introduction of multiple-prism grating oscillators (Kasuya *et al* 1978, Duarte and Piper 1980).

1981–1984: Discovery and refining of multiple-prism grazing-incidence grating cavities (Duarte and Piper 1981, 1984).

1982: Introduction of the generalized multiple-prism grating dispersion theory (Duarte and Piper 1982).

1.2.3 Narrow-linewidth landmarks in CW organic dye lasers

In CW organic dye lasers, where there are no temporal constraints, the laser linewidth varies greatly from system to system and can be approximately in the $0.000\,000\,0009 \leqslant \Delta\lambda \leqslant 0.06$ nm range. At one end is the straight-forward linear mirror-mirror cavity, $\Delta\lambda \approx 0.06$ nm, of Peterson *et al* (1970) and at the other extreme the sophisticated stabilized dye laser, $\Delta\lambda \approx 0.000\,000\,0009$ nm, of Hough *et al* (1987). Very narrow linewidths from stabilized organic dye lasers can be expressed more readily in the frequency domain where linewidths are minimized to $\Delta\nu \approx 750$ Hz (Hough *et al* 1987) and $\Delta\nu \approx 100$ Hz (Drever *et al* 1983). Parenthetically, the stabilization systems developed by Drever and Hall, for the liquid organic CW dye laser, eventually became crucial to the stabilization of second and third generation lasers applied in the detection of *gravitational waves*.

Important landmark developments in this field include:

1970: The CW organic dye laser is discovered (Peterson *et al* 1970).

1973: Frequency stabilized CW dye lasers are introduced (Berger *et al* 1973).

1983: Extremely narrow-linewidth emission in stabilized CW dye lasers is demonstrated (Drever *et al* 1983).

An additional development worth noting in CW organic dye lasers is the demonstration of semiconductor lasers as pump lasers (Scheps 1995).

1.2.4 CW organic dye laser developments for pulse compression

In this subsection, landmark contributions and discoveries pertinent to the development of ultrashort-pulse dye lasers are listed. Ultrashort-pulse lasers, ultrafast lasers, and femtosecond lasers are all homologous lasers developed thanks to the CW organic dye laser. Indeed, it was on the back of the CW organic dye laser that most of the momentous contributions for ultrashort lasers resulted. These discoveries are found applied throughout laser technology, irrespective of gain media, and include:

1967: Mode-locking, using intracavity saturable absorbers, is introduced to the organic dye laser (Schmidt and Schäfer 1968).

1972: Passively mode-locked CW dye lasers are demonstrated (Ippen *et al* 1972).

1976: Introduction of colliding-pulse mode-locking in CW dye lasers (Ruddock and Bradley 1976).

1982–87: Generalized multiple-prism dispersion theory, applicable to pulse compression, is developed (Duarte and Piper 1982, Duarte 1987).

1983–84: Prismatic negative dispersion for pulse compression is demonstrated (Dietel *et al* 1983, Fork *et al* 1984).

1986: Laser pulses as short as 6 fs are reported using multiple-prism and multiple-grating pulse compression (Fork *et al* 1987).

1987: Fiber multiple-prism compressors are demonstrated (Kafka and Baer 1987).

Contemporaneous attosecond pulse lasers and few-cycle pulse lasers have, in one way or another, benefited directly from the physics developed to create femtosecond organic dye lasers. For a detailed review on this subject see Diels (1990) and Diels and Rudolph (1996).

1.3 Solid-state organic lasers

Solid-state organic lasers are a class of lasers that include mainly solid-state organic lasers and semiconductor organic lasers. The excitation of these lasers is performed utilizing optical means, although electrical excitation of coherent organic semiconductor sources has been recently demonstrated.

1.3.1 Solid-state organic dye lasers

In this subsection, important contributions and discoveries pertinent to the development of pulsed tunable solid-state organic dye lasers are listed.

1967: Laser-pumped solid-state dye lasers are discovered (Soffer and McFarland 1967).

1968: Flashlamp-pumped solid-state dye lasers are introduced (Peterson and Snavely 1968).

1994: First tunable narrow-linewidth solid-state dye laser oscillators are demonstrated (Duarte 1994).

1999: Multiple-prism grating solid-state dye laser oscillator demonstrates a linewidth-temporal performance of $\Delta\nu\Delta t \approx 1.06$ (Duarte 1999).
1999–2000: Distributed feedback (DFB) solid-state dye lasers are introduced (Wadsworth *et al* 1999, Zhu *et al* 2000).
2002: Waveguide solid-state dye lasers are introduced (Oki *et al* 2002).

The tunable narrow-linewidth solid-state dye laser oscillators comprise multiple-prism Littrow grating and hybrid multiple-prism grazing-incidence grating configurations (Duarte 1994).

1.3.2 Further notable developments

Here, additional important contributions and discoveries pertinent to the development of optically pumped organic lasers are listed.
1988: Microcavity organic lasers are discovered (De Martini and Jakobovitz 1988).
1990: Organic fiber lasers are discovered (Knobbe *et al* 1990).
1996: Conjugated polymer lasers are announced (Hide *et al* 1996, Holzer *et al* 1996, Tessler *et al* 1996).
1997: Waveguide organic semiconductor lasers are introduced (Kozlov *et al* 1997).
1997: Hybrid waveguide-DFB organic dye lasers are disclosed (Maeda *et al* 1997).
1998: Organic semiconductor vertical-cavity surface–surface emitting lasers (OVCSELs) are introduced (Bulović *et al* 1998).
2006: Optofluidic DFB lasers are disclosed (Psaltis *et al* 2006).

It should be noted that some of these entries are anticipated by US Patents. Detailed reviews discussing various aspects of these developments are given by Kranzelbinder and Leising (2000), Karnutsch (2007), Samuel and Turnbull (2007), and Grivas and Pollnau (2012).

1.3.3 Coherent emission from electrically-pumped organic semiconductors

Spatially coherent and spectrally coherent emission, also characterized as laser emission, from an electrically-pumped organic semiconductor was reported for the first time by Duarte *et al* (2005) and Duarte (2007). This was achieved using a high-luminescence OLED device in series, or tandem, using an active medium of coumarin 545 tetramethyl-doped tris(8-hydroxyquinoline) aluminum (Alq_3). This organic semiconductor dye-doped medium was confined within an integrated interferometric configuration. This subject matter is discussed in detail in chapter 11. In that chapter subsequent claims of coherent emission from electrically pumped organic semiconductors are also examined.

1.4 Organic photonics

Organic photonics is an enormous field that utilizes organic materials to generate, transmit, detect, and process light for a large number of applications in various areas of scientific and technological endeavor. In this regard, it is certainly an improvable task to capture the ineffable essence of the field in the confined space of this book.

A pragmatic approach to confront this limitation, however, is to adopt a rather terse reference-based style to describe the various areas of practical interest that have flourished in this extensive field.

1.5 Organic lasers and organic photonics

Organic Lasers and Organic Photonics is composed of fifteen chapters.

The first group of chapters:

Chapter 2: Organic laser dyes.
Chapter 3: Energetics of organic laser dyes.
Chapter 4: Polymer matrices for lasers.

These chapters include subject matter on organic gain media applicable to organic lasers and organic photonics.

The second group of chapters:

Chapter 5: Cavity and resonator architectures for high-performance organic laser oscillators.
Chapter 6: Mathematical-physics for tunable narrow-linewidth organic laser oscillators.

These chapters concentrate the attention on the architecture and the physics of high-performance organic lasers.

The next chapter:

Chapter 7: Best performance of organic lasers.

This chapter is a handbook style tabulation summarizing the best performance of optically pumped organic lasers as disclosed in the open literature.

The third set of chapters:

Chapter 8: Tunable organic lasers for directed energy.
Chapter 9: Polymer-nanoparticle organic lasers.
Chapter 10: Compact and miniaturized organic dye lasers: from glass to bio-based gain media.
Chapter 11: Electrically-pumped organic semiconductor laser emission.

These chapters focus on specific areas of interest within the larger field of organic sources of coherent radiation.

The last three chapters:

Chapter 12: Organic photonics.
Chapter 13: Organic dyes in optogenetics.

Chapter 14: Tunable organic lasers and organic dyes in medicine.

These chapters focus on use of organic optical media and organic lasers in a variety of utilitarian and exciting applications. In particular, chapters 13 and 14 focus on biomedical applications.

The last chapter:
Chapter 15: Organic lasers for *N*-channel quantum entanglement.

This chapter is a stand-alone chapter that examines the potential of applying organic sources of coherent radiation to quantum communications and quantum entanglement.

1.6 Perspective

Wavelength agility, or tunability, and exquisite high-performance narrow-linewidth emission are two intrinsic characteristics associated with optimized organic lasers. Beyond these characteristics, the field can be divided into large high-energy organic lasers and miniaturized organic lasers. Low capital cost is associated with both these avenues.

More specifically, in regard to optically pumped miniaturized organic lasers: low cost of fabrication and mass production is a major advantage that could have an enormous impact provided these lasers are optimally engineered and optimally coupled to efficient excitation devices such as blue-green diode laser pump sources. Coherent organic integrated interferometric sources directly excited by electrons widen the horizons even further.

Some time ago it was reflected that 'without Maxwell equations and quantum mechanics present day technology including lasers, computers, and the internet, could not have been possible… Quantum mechanics plus electromagnetism provide the bases of the wonderful technologies we enjoy today' (Duarte 2012).

Organic lasers, and organic photonics, are a manifestation of electromagnetism, quantum mechanics, and organic chemistry principles. The emission and transmission of quanta, or photons, play a crucial role in organic lasers and organic photonics. In this regard, the subject matter of this book is not only timely but transcends contemporaneous interests towards the future.

This century has been designated in numerous publications as the 'century of the photon' and that is a most welcome designation. However, the quantum of light, also known as the photon, will certainly dominate a lot more than this century. Photons have been here since the early beginnings … and will be here for aeons to come. It is certainly an immense pleasure, and privilege, to contribute in small part to this brilliant age of discovery via our account of *organic lasers and organic photonics*.

References

Baltakov F N, Barikhin B A and Sukhanov L V 1974 400 J pulsed laser using a solution of rhodamine 6 G in ethyl alcohol *JETP Lett.* **19** 174–5

Barger R L, Sorem M S and Hall J L 1973 Frequency stabilization of a CW dye laser *Appl. Phys. Lett.* **22** 573–575
Bass I L, Bonano R E, Hackel R H and Hammond P R 1992 High-average-power dye laser at Lawrence Livermore National Laboratory *Appl. Opt.* **31** 6993–7006
Bulović V, Kozlov V G, Khalfin V B and Forrest S R 1998 Transform-limited narrow-linewidth lasing action in organic semiconductor microcavities *Science* **279** 553–5
De Martini F and Jakobovitz J R 1988 Anomalous spontaneous-emission-decay phase transition and zero-threshold laser action in a microscopic cavity *Phys. Rev. Lett.* **60** 1711–4
Diels J-C 1990 Femtosecond dye lasers *Dye Laser Principles* ed F J Duarte and L W Hillman (New York: Academic) ch 3
Diels J-C and Rudolph W 1996 *Ultrafast Laser Pulse Phenomena* (New York: Academic)
Dietel W, Fontaine J J and Diels J-C 1983 Intracavity pulse compression with glass: a new method of generating pulses shorter than 60 fs *Opt. Lett.* **8** 4–6
Drever R W P, Hall J L, Kowalski F V, Hough J, Ford G M, Munley A J and Ward H 1983 Laser phase and frequency stabilization using an optical resonator *App. Phys.* B **31** 97–105
Duarte F J 1987 Generalized multiple-prism dispersion theory for pulse compression in ultrafast dye lasers *Opt. Quantum Electron.* **19** 223–9
Duarte F J (ed) 1991 *High-Power Dye lasers* (Berlin: Springer)
Duarte F J 1992 *Selected Papers on Dye Lasers* (Bellingham, WA: SPIE)
Duarte F J 1994 Solid-state multiple-prism grating dye-laser oscillators *Appl. Opt.* **33** 3857–60
Duarte F J 1999 Multiple-prism grating solid-state dye laser oscillator: optimized architecture *Appl. Opt.* **38** 6347–9
Duarte F J 2007 Coherent electrically excited organic semiconductors: visibility of interferograms and emission linewidth *Opt. Lett.* **32** 412–4
Duarte F J 2012 Unpublished.
Duarte F J and Hillman L W (ed) 1990 *Dye Laser Principles* (New York: Academic)
Duarte F J, Liao L S and Vaeth K M 2005 Coherence characteristics of electrically excited tandem organic light-emitting diodes *Opt. Lett.* **30** 3072–4
Duarte F J and Piper J A 1980 A double-prism beam expander for pulsed dye lasers *Opt. Commun.* **35** 100–4
Duarte F J and Piper J A 1981 Prism preexpanded grazing-incidence grating cavity for pulsed dye lasers *Appl. Opt.* **20** 2113–6
Duarte F J and Piper J A 1982 Dispersion theory of multiple-prism beam expander for pulsed dye lasers *Opt. Commun.* **43** 303–7
Duarte F J and Piper J A 1984 Narrow-linewidth high-prf copper laser-pumped dye laser oscillators *Appl. Opt.* **23** 1391–4
Fork R L, Brito Cruz C H, Becker P C and Shank C V 1987 Compression of optical pulses to six femtoseconds by using cubic phase compensation *Opt. Lett.* **12** 483–5
Fork R L, Greene B I and Shank C V 1981 Generation of optical pulses shorter than 0.1 psec by colliding pulse mode locking *Appl. Phys. Lett.* **38** 671–2
Fork R L, Martinez O M and Gordon J P 1984 Negative dispersion using pairs of prisms *Opt. Lett.* **9** 150–2
Grivas C and Pollnau M 2012 Organic solid-state integrated amplifiers and lasers *Lasers Photon, Rev.* **6** 419–62
Hänsch T W 1972 Repetitively pulsed tunable dye laser for high-resolution spectroscopy *Appl. Opt.* **11** 895–8

Hide F, Schwartz B J, Díaz-García M and Heeger A L 1996 Laser emission from solutions and films containing polymer and titanium dioxide nanocrystals *Chem. Phys. Lett.* **256** 424–30
Holzer W, Penzkofer A, Gong S-H, Bleyer A and Bradley D D C 1996 Laser action in poly (m-phenylenevinylene-co-2, 5-dioctoxy-p-phenylenevinylene) *Adv. Mat.* **8** 974–8
Hough J, Hils D, Rayman M D, Ma L-S, Hollberg L and Hall J L 1987 Dye-laser frequency stabilization using optical resonators *Appl. Phys.* B **33** 179–85
Ippen E P, Shank C V and Dienes A 1972 Passive mode locking of the CW dye Laser *Appl. Phys. Lett.* **21** 348–50
Kafka J D and Baer T 1987 Prism-pair dispersive delay lines in optical pulse compression *Opt. Lett.* **12** 401–3
Karnutsch C 2007 *Low Threshold Organic Thin Film Laser Devices* (Göttingen: Cuvillier)
Kasuya T, Suzuki T and Shimoda K 1978 A prism anamorphic system for Gaussian beam expander *Appl. Phys.* **17** 135–6
Knobbe E T, Dunn B, Fuqua P D and Nishida F 1990 Laser behavior and photostability characteristics of organic dye doped silicate gel materials *Appl. Opt.* **29** 2729–33
Kogelnick H and Shank C V 1971 Stimulated emission in a periodic structure *Appl. Phys. Lett.* **18** 152–4
Kozlov V G, Bulović V, Burrows P E and Forrest S R 1997 Laser action in organic semiconductor waveguide and double-heterostructure devices *Nature* **389** 362–4
Kranzelbinder G and Leising G 2000 Organic solid-state lasers *Rep. Prog. Phys.* **63** 729–62
Littman M G and Metcalf H J 1978 Spectrally narrow pulsed dye laser without beam expander *Appl. Opt.* **17** 2224–7
Maeda M 1984 *Laser Dyes* (New York: Academic)
Maeda M, Oki Y and Imamura K 1997 Utrashort pulse generation from an integrated single-chip dye laser *IEEE J. Quantum Electron.* **33** 2146–9
Oki Y, Aso K, Zuo D, Vasa N J and Maeda M 2002 Wide-wavelength range operation of a distributed-feedback dye laser with a plastic waveguide Japan *J. Appl. Phys.* **41** 6370–6374
Peterson O G and Snavely B B 1968 Stimulated emission from a flashlamp-excited organic dyes in polymethyl mehacrylate *Appl. Phys. Lett.* **12** 238–40
Peterson O G, Tuccio S A and Snavely B B 1970 CW operation of an organic dye solution laser *Appl. Phys. Lett.* **17** 245–7
Psaltis D, Quake S R and Yang C 2006 Developing optofluidic technology through the fusion of microfluidics and optics *Nature* **442** 381–6
Ruddock I S and Bradley D J 1976 Bandwidth-limited subpicosecond pulse generation in mode-locked CW dye lasers *Appl. Phys. Lett.* **29** 296–7
Saikan S 1978 Nitrogen-laser-pumped single-mode dye laser *Appl. Phys.* **17** 41–4
Samuel I D W and Turnbull G A 2007 Organic semiconductor lasers *Chem. Rev.* **107** 1272–95
Scheps R 1995 Near-IR dye laser for diode-pumped operation *IEEE J. Quantum Electron.* **31** 126–34
Schäfer F P (ed) 1990 *Dye Lasers* 3rd ed (Berlin: Springer)
Schäfer F P, Schmidt W and Volze J 1966 Organic dye solution laser *Appl. Phys. Lett.* **9** 306–9
Schmidt W and Schäfer F P 1968 Self-mode-locking of dye-laser with saturable absorbers *Phys. Lett.* **26** 558–9
Shohan I, Danon N N and Oppenheim U P 1977 Narrow band operation of pulsed dye laser without beam expansion *J. Appl. Phys.* **48** 4495–7

Soffer B H and McFarland 1967 Continuously tunable narrow-band organic dye lasers *Appl. Phys.* **10** 266–7

Sorokin P P and Lankard J R 1966 Stimulated emission observed from an organic dye, chloro-aluminum phthalocyanine *IBM J. Res. Dev.* **10** 162–3

Spaeth M L and Bortfeld D P 1966 Stimulated emission from polymethine dyes *Appl. Phys.* **9** 179–81

Tang K Y, O'Keefe T, Treacy B, Rottler R and White C 1987 Kilojoule-output XeCl dye laser: optimization and analysis *Proc. Dye Laser/Laser Dye Technical Exchange Meeting 1987* ed J H Bentley (Alabama: U. S. Army Missile Command, Redstone Arsenal), pp 490–502

Tessler N, Denton G J and Friend R H 1996 Lasing from conjugated polymer microcavities *Nature* **382** 695–7

Wadsworth W J, McKinnie I T, Woolhouse A D and Haskell T G 1999 Efficient distributed feedback solid state dye laser with dynamic grating *Appl. Phys.* B **69** 163–5

Zhu X-L, Lam S-K and Lo D 2000 Distributed-feedback dye-doped solgel silica lasers *Appl. Opt.* **39** 3104–7