# Solvent-free fluidic organic dye lasers

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**Abstract:** We report on the demonstration of liquid organic dye lasers based on 9-(2-ethylhexyl)carbazole (EHCz), so-called liquid carbazole, doped with green- and red-emitting laser dyes. Both waveguide and Fabry-Perot type microcavity fluidic organic dye lasers were prepared by capillary action under solvent-free conditions. Cascade Förster-type energy transfer processes from liquid carbazole to laser dyes were employed to achieve color-variable amplified spontaneous emission and lasing. Overall, this study provides the first step towards the development of solvent-free fluidic organic semiconducting lasers and demonstrates a new kind of optoelectronic applications for liquid organic semiconductors.

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## 1. Introduction

In recent years, liquid organic semiconductors have emerged as a promising class of materials for organic optoelectronics [1–3]. These soft optoelectronic functional materials present several advantages over conventional organic semiconductors, including solvent-free device processing, ultimate mechanical flexibility and tunable optoelectronic responses. Organic conjugated molecules which are liquid at room temperature have been successfully used in a variety of organic optoelectronic applications such as photorefractive devices [4], organic light-emitting diodes (OLEDs) [5–7], dye-sensitized solar cells [8] and bistable memories [9]. Microfluidic OLEDs were recently reported in which fresh liquid organic semiconductors could be continuously supplied to the emitting layer [7]. Such a convectional circulation of a liquid organic semiconducting material can be applied to the development of degradation-free organic optoelectronic devices. In that context, the demonstration of solvent-free optofluidic semiconducting organic lasers would be an important step towards the development of liquid organic optoelectronic applications.

Over the past few years, optofluidic dye lasers have gained interest as miniature coherent light sources for integrated optics and lab-on-a-chip applications [10]. In particular, these devices have been shown to be very promising for high sensitivity chemical and biosensing [11]. To date, a variety of optical resonator structures have been used for these microfabricated laser sources including Fabry-Perot cavities, distributed feedback gratings, microdroplets and microring cavities [12–15]. The solution of organic dye molecules injected into the optofluidic dye lasers provides the optical gain and thus plays a critical role on the optical properties of the laser emission. However, up to now, these liquid dye solutions have

been always prepared in a solvent and thus do not show any semiconducting properties. Here, we demonstrate that liquid organic semiconducting materials can be used as gain medium in solvent-free optofluidic organic lasers [16–18] allying the nonvolatility and the high concentration of electronically active π-conjugated moieties typically obtained in organic semiconducting films [19] with the versatility and tunability of liquid dye lasers [20].

In this paper, we investigate the amplified spontaneous emission (ASE) and lasing properties of 9-(2-ethylhexyl)carbazole (EHCz) mixed with coumarin 153 (C153) and [2-[2-[4-(dimethylamino)phenyl]ethenyl]-6-methyl-4H-pyran-4-ylidene]-propanedinitrile (DCM). Previous work has shown that EHCz, so-called liquid carbazole, is liquid at room temperature with a glass transition temperature well below 0°C [1]. This liquid organic semiconductor is a blue fluorescent emitter, exhibits good hole transport properties and has been successfully used as a liquid hole transport matrix for rubrene in OLEDs [5] and silver nanoparticles in bistable memories [9]. In this work, we incorporate liquid dye-doped EHCz blends into two different optofluidic device geometries based on waveguide [21] and Fabry-Perot type microcavity [22] structures, respectively. Cascade energy transfer from the liquid carbazole to green and red-emitting laser dyes is used to tune effectively the ASE and laser emission [23]. The results provide evidence that liquid carbazole can be used as a functional emissive matrix for optofluidic laser applications. Accordingly, the possibility to integrate liquid organic semiconductors with tunable optoelectronic properties into solvent-free optofluidic devices opens new alternatives for the next generation of liquid organic optoelectronic devices.
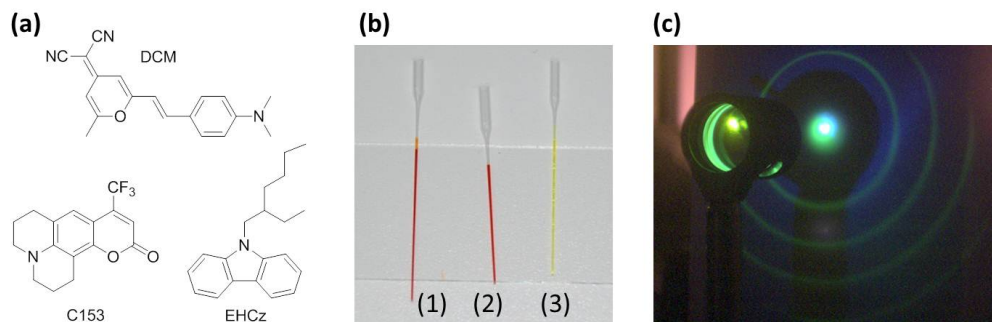


Fig. 1. (a) Chemical structure of liquid carbazole (EHCz), coumarin 153 (C153) and DCM. (b) Photographs of waveguiding capillary tubes filled with (1) EHCz:C153:DCM (95; 3.1; 1.9 wt.%), (2) EHCz:DCM (97.5:2.5 wt.%) and (3) EHCz:C153 (96.5: 3.5 wt.%). (c) Photograph of interference fringes observed on a screen from a Fabry-Perot type microcavity solvent-free liquid organic green dye laser filled with EHCz:C153 (96.5; 3.5 wt.%). The device was optically pumped at 355 nm by the third harmonic of a nanosecond Nd:YAG laser.

## 2. Experimental methods

The chemical structures of EHCz, C153 and DCM are shown in Fig. 1(a). These organic materials were purchased from Sigma-Aldrich and used without further purification. The laser dyes were mixed with liquid carbazole at various concentrations in chloroform solution. After the full evaporation of the residual solvent, the liquid composite materials were incorporated into optofluidic devices by capillarity. Figures 1(b) and 1(c) show photographs of waveguiding and Fabry-Perot type microcavity devices filled with the liquid carbazole blends.

Absorption and fluorescence spectra were recorded using an absorption spectrophotometer (Hitachi U-3310) and a steady-state fluorimeter (PTI QuantaMaster 40) respectively. For the fluorescence spectra measurements, the liquid emitting material was sandwiched between two identical precleaned fused silica substrates. For the absorption spectra measurements in solution, the molecules were dissolved in chloroform and the solutions were placed in a 1cm quartz cuvette.

Single core capillary glass tubes used in this work for making waveguide liquid organic light-emitting devices had a core diameter and a glass cladding thickness of $500 \pm 100$ and 10 µm respectively. Due to the low viscosity of liquid carbazole at room temperature, the infilling process of these tubes by capillary action was carried out by directly dipping them in the liquid organic composite materials. The other type of optofluidic organic laser devices was based on Fabry-Perot type microcavities. The liquid blends were placed between two identical dielectric mirrors with a high reflectance at the emission wavelengths of the green and red organic lasers (Melles Griot, model AR1-1025-C-0 with a reflectance R > 98% between 470 and 550 nm, model HN-1025-C-0 with a reflectance R > 98% between 600 and 680 nm). The thickness of the Fabry-Perot cavities was controlled using silica spheres with a diameter of 5 µm dispersed into a silicon paste.

The fluidic organic devices were optically pumped at 355 nm using the third harmonic of a Nd:YAG laser (Continuum SureLite II) with a repetition rate and pulse duration of 10 Hz and 6-8 ns, respectively. The waveguiding structures were excited perpendicularly to the long axis of the capillary tubes by a stripe of dimension 0.15 mm x 1 cm. The axial emission from the liquid organic materials was collected at one end of the capillary tubes using an optical fiber coupled to a CCD spectrometer (Avantes Avaspec 2048). For the characterization of the microcavity devices, the diameter of the excitation laser beam illuminating the samples was 3 mm and the incidence angle was around 10°. The laser output was collected perpendicularly to the microcavities also using the optical fiber coupled to the CCD spectrometer. Note that all the preparation and characterization of the devices were performed in this work under ambient atmosphere conditions.
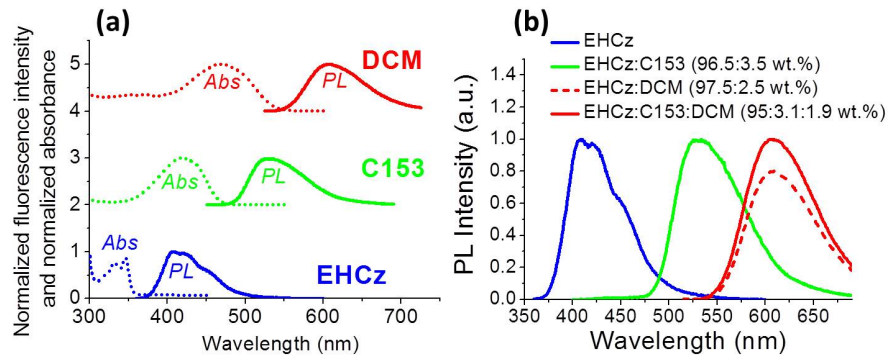


Fig. 2. (a) Normalized absorbance and photoluminescence spectra of the liquid host material and the laser dyes. The spectra from EHCz are measured from a solvent-free neat layer whereas the other spectra are from solutions in chloroform. Note that the spectra measured in C153 and DCM are vertically shifted for clarity reasons. (b) Photoluminescence spectra measured in solvent-free liquid carbazole blends. The excitation wavelength was 350 nm.

## 3. Energy transfer from liquid carbazole to laser dyes

The absorption and fluorescence spectra of a neat liquid EHCz layer sandwiched between two fused silica substrates are shown in Fig. 2(a). It can be seen that liquid carbazole is fully transparent in the visible and strongly absorbs light in the ultraviolet. The two peaks observed at 335 and 348 nm are assigned to the absorption from the carbazole monomer unit [1]. The fluorescence spectrum of EHCz shows a blue emission with a maximum at 410 nm. Figure 2(a) also shows the absorption and emission spectra of C153 and DCM in chloroform. The maximum in the fluorescence spectrum of these dyes is observed at 533 and 609 nm respectively. It is well-known that Förster-type energy transfer depends on the overlap between the emission spectrum of the donor molecule and the absorption spectrum of the acceptor [23–26]. The overlaps observed between the optical spectra shown in Fig. 2(a) suggest that energy transfer can take place from EHCz to C153 and DCM as well as from

C153 to DCM. Note that energy transfer from carbazole derivatives to coumarin dyes have been successfully used in OLEDs and organic solid-state lasers [27,28].

We prepared three different solvent-free liquid blends: EHCz:C153 (96.5:3.5 wt.%), EHCz:DCM (97.5:2.5 wt.%) and EHCz:C153:DCM (95:3.1:1.9 wt.%) and placed them between two fused silica substrates. The two laser dyes were well dissolved in the liquid carbazole host and our samples presented an excellent optical homogeneity. The fluorescence spectra of the three blends, which are displayed in Fig. 2(b), were measured using an excitation wavelength of 350 nm. At such a wavelength, the absorption of the blends is dominated by the contribution from the liquid carbazole host. The emission spectra provide evidence that energy transfer takes place from the liquid carbazole matrix to the C153 and DCM dyes in good consistency with the resonance energy transfer theory [29]. We also found that the ternary blend (EHCz:C153:DCM) shows a similar fluorescence spectrum as the binary blend (EHCz:DCM). However, the fluorescence efficiency from the DCM dyes is significantly increased by the incorporation of C153, which can be attributed to cascade energy transfer from EHCz to DCM through C153 [23].
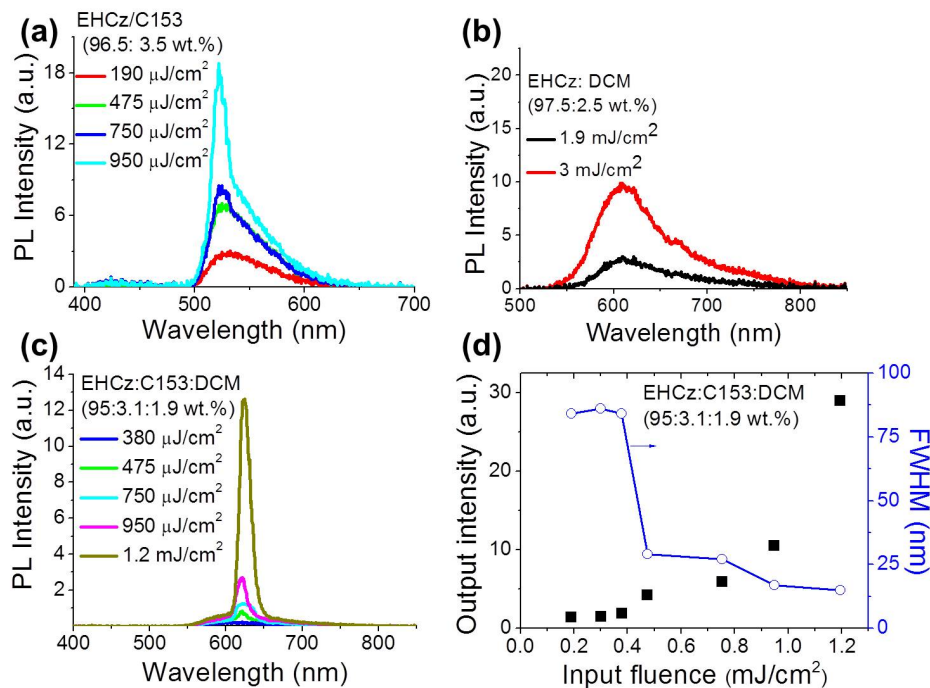


Fig. 3. Emission spectra measured in single core waveguiding capillary tubes filled with liquid carbazole blends at different pumping intensities. The excitation wavelength was 355 nm. The three blends used in these measurements are: (a) EHCz:C153 (96.5: 3.5 wt.%), (b) EHCz:DCM (97.5: 2.5 wt.%) and (c)) EHCz:C153:DCM (95: 3.1: 1.9 wt.%). (d) Output intensity and full width at half maximum (FWHM) of the ASE spectra as a function of the input fluence in waveguiding devices filled with EHCz:C153:DCM (95: 3.1: 1.9 wt.%).

## 4. Amplified spontaneous emission and lasing in solvent-free dye-doped liquid carbazole

Single-core capillary tubes were filled with solvent-free liquid carbazole blends EHCz:C153 (96.5:3.5 wt.%), EHCz:DCM (97.5:2.5 wt.%) and EHCz:C153:DCM (95:3.1:1.9 wt.%). Note that the refractive index of liquid carbazole is around 1.7 and thus is higher than that of the glass of the capillary tubes. The waveguiding structures were pumped by nanosecond pulses at 355 nm and their axial emission spectra were recorded for various pump intensities. At low pump intensities, their emission spectra remain identical to the spontaneous emission spectra

displayed in Fig. 2(b). However, as shown in Fig. 3, the emission from EHCz:C153 and EHCz:C153:DCM become narrower at high excitation densities. We attribute this line narrowing to amplified spontaneous emission (ASE) [23,24,30–32], which occurs when spontaneously emitted photons are waveguided and amplified in the gain medium by stimulated emission due to a population inversion. Amplification in the capillaries filled with EHCz:C153 and EHCz:C153:DCM is clearly observed at 522 and 625 nm, respectively. The excitation density characterizing the ASE threshold in EHCz:C153 is around 500-600 $\mu$J/cm$^2$ and the full-width at half maximum (FWHM) of its emission spectrum drops from 82 to 15 nm as the pumping intensity is increased from 190 to 950 $\mu$J/cm$^2$. In contrast, no ASE peak was observed in EHCz:DCM even at pumping fluence as high as 3 mJ/cm$^2$. This is certainly due to the fact that energy transfer from EHCZ to DCM is less efficient than that from EHCz to C153. As shown in Fig. 3(d), adding an appropriate amount of C153 to EHCz:DCM leads to a lower ASE threshold in the range of 500-600 $\mu$J/cm$^2$, which is comparable to that measured in EHCz:C153. It can also be seen that the FWHM of the EHCZ:C153:DCM emission spectrum decreases from 85 to 15 nm as the excitation density is increased. These results demonstrate the interest in using cascade energy transfer for tuning efficiently over a broad region of wavelengths the lasing properties of solvent-free liquid organic semiconducting gain media. It should be pointed out here that whispering gallery mode (WGM) lasing has been previously observed in dye-doped solvent flowing in a silica capillary fiber [33]. It is possible that such a WGM lasing is emitted from our optofluidic waveguiding devices. These whispering gallery modes typically show very narrow spacing modes and should be identified using a spectrophotometer with a higher spectral resolution than that used in our experiment.
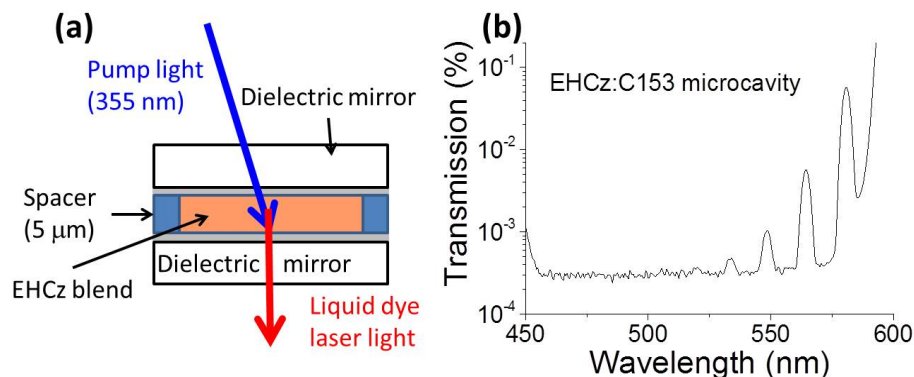


Fig. 4. (a) Schematic representation of a Fabry-Perot type microcavity solvent-free fluidic organic dye laser. (b) An example of transmission spectrum measured in a microcavity filled with EHCz:C153 (96.8:3.2 wt.%) using unpolarized white light at normal incidence. For this sample, four cavity modes were clearly observed at 533, 548, 564 and 580 nm.

Figure 4(a) displays a schematic representation of Fabry-Perot type microcavity organic dye lasers used in this study. The green- and red-light-emitting devices were filled with EHCz:C153 (96.8:3.2 wt.%) and EHCz:C153:DCM (95:3.1:1.9 wt.%) respectively. The transmission spectrum of a EHCz:C153 microcavity is shown in Fig. 4(b) and was obtained using an unpolarized white light at normal incidence. For this specific sample, four cavity modes were observed in the spectrum at 533, 548, 564 and 580 nm with a FWHM varying between 3 and 4 nm. This leads to a quality factor Q ranging from 130 to 160. From the position of the modes in the transmission spectrum of Fig. 4(b), the physical thickness of the microcavity is found to be around 5.8 ± 0.2 $\mu$m.
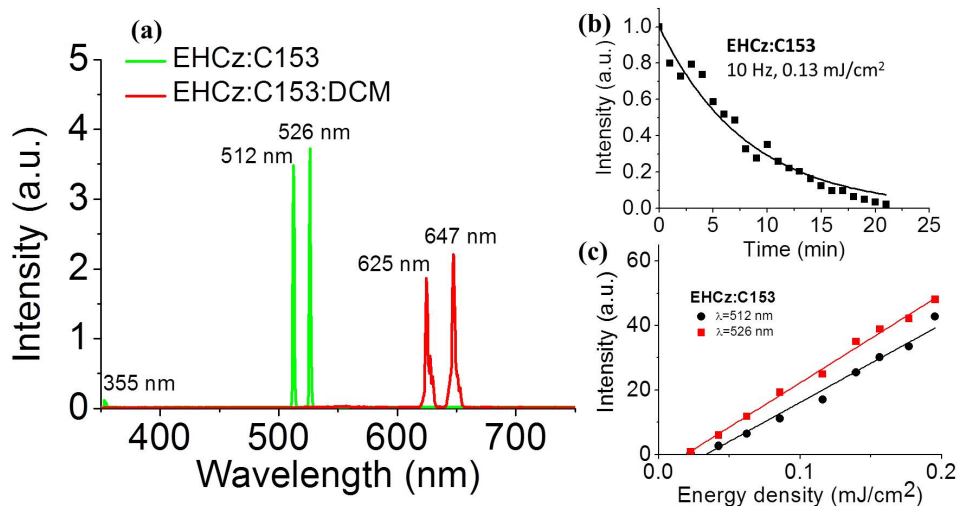
Fig. 5. (a) Laser emission spectra from Fabry-Perot type solvent-free liquid organic microcavities based on EHCz:C153 (96.8:3.2 wt.%) and EHCz:C153:DCM (95:3.1:1.9 wt.%). (b) Temporal decay of the laser emission intensity from the EHCz:C153 microcavity for pulse repetition rate of 10 Hz and a pumping energy density of about 0.13 mJ/cm$^2$. The solid line corresponds to a fit by a single exponential decay function. (c) Output intensity as a function of the input energy density showing a laser emission with a lasing threshold for wavelengths of 512 and 526 nm from the EHCz:C153 microcavity laser.

As shown in Fig. 5(a), above the laser threshold, dual wavelength laser emission was observed in both devices. Green laser emission at 512 and 526 nm was observed in the emission spectrum of EHCz:C153 (96.8:3.2 wt.%). In the case of EHCz:C153:DCM (95:3.1:1.9 wt.%), the red laser emission peaks were located at 625 and 647 nm. The FWHM of the laser modes was found to be around 1 nm, which is the resolution limit of our spectrometer. This dual wavelength laser emission is presumably due to the large pump spot size area used in this work. Such a pumping configuration can lead to resonance conditions changing over the excited area and thus to multi-mode operation [34]. However, degradation of the organic layer is reduced and the device stability during our measurements was improved. As displayed in Fig. 5(b), we monitored the decay of the laser emission from the EHCz:C153 microcavity for a 355 nm pumping energy density of about 0.13 mJ/cm$^2$ at a repetition rate of 10 Hz. The decay can be fitted by a single exponential function with an average characteristic response time of about 10 minutes. This response time was found to decrease as the pumping beam spot size is reduced or the pumping energy density is increased. Note that an extinction of the laser emission was typically observed after 30-60 minutes of continuous operation depending on the pump power. This decay can be explained by bleaching of the light-emitting molecules. Because the viscosity of EHCz is higher than solvents commonly used in optofluidics, the bleached molecules cannot be replaced by diffusion only [35] and the bleaching should be compensated by externally pumping the liquid medium for more stable continuous operation of the devices.

Figure 5(c) shows the output emission intensity as a function of the excitation energy density for the two laser lines observed from EHCz:C153 (96.8:3.2 wt.%). The laser threshold energy density was found to be as low as 33 and 19 μJ/cm$^2$ for the emission at 512 and 526 nm, respectively. Such low lasing threshold values are on the same order of magnitude as those measured in dye-doped organic solid-state distributed feedback lasers using 4,4'-N,N'-dicarbazole-1,1'-biphenyl (CBP) as a host, which presented laser threshold energy densities ranging from 7.5 to about 70 μJ/cm$^2$ [36]. In case of the EHCz:C153:DCM microcavity, the threshold values were higher and close to 480 μJ/cm$^2$. It is worth noting here that a weak fluorescence from C153 could be observed in this specific device, which indicates that some

excitons formed on C153 were not transferred to DCM before their radiative decay. Although the blend composition would need to be optimized to reduce the fluorescence from C153 and to improve the cascade energy transfer process, our lasing threshold energy density values are lower than that of about 2.5 mJ/cm$^2$ measured in a wedge-shaped Fabry-Perot type microcavity organic laser based on a tris(8-hydroxy quinoline) aluminum (Alq$_3$): DCM blended film with a thickness varying from 180 to 1850 nm [34]. Note that this previous work used a small excitation spot size of about 10 microns and that their threshold value expressed in nJ/pulse was found to be as low as 2 nJ/pulse.

Overall, this work demonstrates solvent-free liquid organic semiconducting dye lasers and the performances of these devices are already comparable with those typically obtained in organic solid-state dye lasers.

## 4. Conclusions

In summary, we examined the amplified spontaneous emission and lasing properties of the liquid carbazole EHCz blended with the laser dyes Coumarin 153 and DCM. Waveguide and Fabry-Perot type microcavity organic dye laser devices were prepared by capillary action under solvent-free conditions. The practicality of these laser sources is enhanced by the possibility of tuning effectively their emission via a cascade energy transfer scheme. This study demonstrates that liquid organic conjugated light-emitting materials can be used as gain medium for the realization of tunable solvent-free fluidic organic lasers and paves the way for the development of novel optofluidic systems with semiconducting functionalities.