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# Modeling of threshold and dynamics behavior of organic nanostructured lasers

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## Abstract

Organic dye molecules offer significant potential as gain media in the emerging field of optical amplification and lasing at subwavelength scales. Here, we investigate the laser dynamics in systems comprising subwavelength-structured cavities that incorporate organic dyes. To this end, we have developed a comprehensive theoretical framework able to accurately describe the interaction of organic molecules with any arbitrary photonic structure to produce single-mode lasing. The model provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers, and also the duration over which lasing action can be sustained before the dye photobleaches. Both the physical properties of the dyes and the optical properties of the cavities are considered. We also systematically studied the feasibility of achieving lasing action under continuous-wave excitation in optically pumped monolithic organic dye lasers. This study suggests routes to realize an organic laser that can potentially lase with a threshold of only a few  $W/cm^2$ . Our work puts forward a theoretical formalism that could enable the advancement of nanostructured organic-based light emitting and sensing devices.

## I. INTRODUCTION

Organic dye lasers with high tunability in the visible wavelengths have attracted interest for many years due to their low-cost processing, flexible choice of substrates, and large emission cross sections that can cover the spectral region from ultraviolet to the near infrared. Electrically pumped organic semiconductor lasers are desirable from a practical standpoint but their realizations have remained elusive due to their high thresholds, low charge-carrier mobility in organic materials, and efficient exciton annihilation process in solid-state organic media<sup>1-5</sup>. A compromise to the above is the hybrid electrically pumped organic lasers<sup>6,7</sup>. These lasers are optically pumped by small electrically driven inorganic diode lasers and exhibit the benefits of a direct electrical pumping scheme (i.e. voltage/current controlled), thus reducing the size and the cost of the polymer laser system. While optically pumped organic lasers have been widely demonstrated<sup>8-11</sup>, lasing is only possible with high peak power excitation sources of short pulses; no continuous-wave (CW) operated organic lasers has been demonstrated without liquid dye circulation.

The motivation for this work is to explore the feasibility of achieving low-threshold lasing in an optically pumped CW monolithic organic laser, where the organic medium is stationary with respect to the pump beam and lasing action is sustained over a time scale longer than all decay lifetimes of the dye. To achieve this, one would need to (*i*) lower the thresholds of existing organic lasers to levels that are commensurate with most commercial applications (typically few  $W/cm^2$ )<sup>12</sup>, (*ii*) minimize the photobleaching effects in the organic media, and (*iii*) ensure thermal management of the devices. In the following, we assume that thermal effects produced by localized heating of the dyes under excitations are minimized over the lasing duration (e.g. few tens of milliseconds in a non-flowing dye laser<sup>13</sup>) through a careful design of the device<sup>14</sup>. We focus instead on optimizing the physical properties of the dyes and optical properties of the cavities to achieve (*i*) and (*ii*), assuming a photobleaching rate determined by the system chemical composition of the organic molecule, the host matrix, and packaging. To do so, it is critical to develop a complete theoretical understanding of the physical mechanisms underlying lasing action in organic lasers.

Here, we introduce a theoretical framework that extends previous studies on organic dyes<sup>15,16</sup> in order to account for the full multi-level rate equations, the photobleaching of dyes, and the modification that a complex electromagnetic environment introduces to the radiative decay rates of the dyes with respect to their free space values (the so-called Purcell effect<sup>17,18</sup>). The formalism

provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers, as well as the duration over which lasing action can be sustained before the dye photobleaches. In particular, using a solution of Rhodamine 6G (R6G) dye molecules as the gain medium, our model suggests routes to attain a low-threshold organic laser that can potentially lase at a few  $W/cm^2$ . We expect our findings to open a path to the realization of cheap and compact organic light sources based on the hybrid electrical pumping scheme, which will in turn open up several potential applications in the visible spectrum. Our results are also of importance in the context of loss-compensation and lasing action in nanophotonics, plasmonics, and metamaterials, where solutions of R6G are often incorporated as the active medium in the system<sup>19,20</sup>.

The paper is organized as follows: Section II (and Appendix A) discuss the general theoretical model. Section II C focuses on lasing action in organic lasers under pulse excitations, where results predicted by the theoretical model have been verified with measurements. Section III presents a detailed analysis of organic lasers under CW excitations. Finally, in Sec. IV, we summarize our results and provide a set of conclusions.

## II. THEORETICAL FRAMEWORK

As a general energy-level model of an organic molecule, we consider the three lowest singlet and two lowest triplet states. Figure 1(a) illustrates the energy level diagram and the different energy transfer mechanisms that form the basic structure of our model. Often, a four-level gain model of only the two lowest singlet states ( $S_0$  and  $S_1$ ) suffices in analyzing systems where the influences of the triplet states are minimized, either by pulsed excitations with widths much shorter than the intersystem crossing lifetimes<sup>21</sup> or by fast circulation of the dye solutions<sup>14,22</sup>. In this work, we considered the full energy-level model shown in Fig. 1(a) and (b) to identify the key parameters (from both molecular and optical standpoints) that can be optimized in order to simultaneously decrease the thresholds, increase the slope efficiencies, and reduce the photobleaching rates under CW pumping in monolithic organic lasers. We would like to point out that while the model described here represents fluorescing molecules dispersed in amorphous environment, it can be simply modified to other configurations such as solid-state Alq<sub>3</sub>:DCM (based on Förster energy transfer) and crystalline organic molecules by modifying the model of the energy levels. In this work, the model is applied to the case of optically-pumped lasing systems consisting of R6G molecules dissolved in a solution under CW and pulsed excitations.

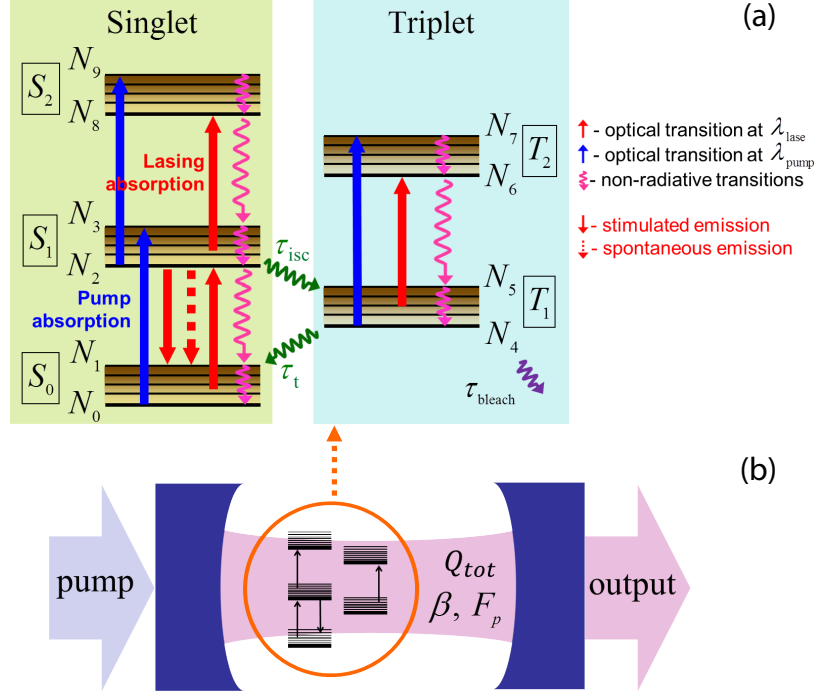


FIG. 1: Laser system consisting of an organic active medium embedded in a passive cavity. (a) The energy-level structure of the considered organic medium comprises of the three lowest singlet states of  $S_0$ ,  $S_1$ , and  $S_2$  and the two lowest triplet states of  $T_1$  and  $T_2$ . Red (blue) solid vertical arrows are optically-induced transitions at the lasing (excitation) wavelength whereas the dashed vertical arrow is the radiative decay of molecules (spontaneous emission). Wavy arrows are non-radiative transitions. Intersystem crossing of molecules from the singlet to triplet states is characterized by the decay lifetime  $\tau_{isc}$  while the subsequent decay of triplets back to the ground singlet state is characterized by  $\tau_t$ . Photobleaching in the organic medium is modeled as the loss of molecules from the triplet state at a rate of  $1/\tau_{bleach}$  (represented as a violet wavy arrow). (b) Optical properties of the laser cavity are the spontaneous emission modification factor  $F_p$ , spontaneous emission coupling factor  $\beta$ , and the total quality factor  $Q_{tot}$  (see text for details on the definitions of these magnitudes).

The various processes in the gain model are described via rate equations (see Appendix A) which are presented in a form that is convenient for numerical analysis, and yet allow for direct relations to experimental parameters. The most general description of an energy level model includes terms representing the optically-induced transitions that are characterized by their absorption and emission cross-sections, as well as the radiative and non-radiative relaxations that

are characterized by their lifetimes. Optically-induced transitions of molecules to the upper states act as resonant absorbers of the excitation and emitted light, while those to the lower states result in the stimulated emission of light, producing lasing photons when the pump power is above the threshold. On the other hand, radiative relaxation of molecules to the ground singlet state leads to the spontaneous emission of light. The non-radiative processes considered are the internal conversions due to rotational and vibrational relaxations, the intersystem crossing of the molecules from the excited singlet state  $S_1$  to the triplet state  $T_1$ , as well as their subsequent de-excitations back to the ground state  $S_0$ . Photobleaching is also included in our analysis and is modeled as a loss of molecules in the triplet state at a rate that is dependent on the excitation conditions<sup>23–25</sup> (i.e. pump power and the presence of oxygen).

Build-up of the triplet states' population can occur when  $T_1$  is energetically closer to  $S_1$  than  $S_0$ , resulting in the transition rate between  $S_1$  and  $T_1$  to be faster than that between  $T_1$  and  $S_0$ . High occupancies in the triplet states have three detrimental effects on the lasing action: the population inversion becomes limited due to the smaller number of molecules in the singlet states, the fraction of the excitation light absorbed by the triplet states is increased, and dye absorption of the stimulated light is enhanced due to the overlapping emission and triplet-absorption spectrum. To overcome these effects, in organic dye solutions for example, quenchers can be used to decrease the decay lifetime of the triplets<sup>26,27</sup>. Our model allows to study the effect of the intersystem crossing lifetime ratio ( $\tau_t/\tau_{isc}$ ) on the triplet excited-state absorption.

The organic gain model is coupled to the semiclassical rate equation describing the population of the lasing photons. These photons are produced by stimulated and spontaneous emission, and at the same time, are resonantly reabsorbed by the organic media. In addition, the photons within the laser cavity are lost due to radiation into the far-field, intrinsic material absorption, and scattering from fabrication disorders in actual devices.

Using the organic laser model of Fig. 1 and the notations summarized in Table I of Appendix B, lasing action is obtained as follows: dye molecules are primarily excited from  $N_0$  to  $N_3$  at a rate proportional to the pump power  $P_{src}$ . The excited molecules then decay nonradiatively into  $N_2$  after a lifetime  $\tau_{32}$ . A net decay of molecules from  $N_2$  to  $N_1$  occurs through stimulated and spontaneous emission, as well as nonradiative relaxation. Some of the molecules in  $N_2$ , however, decay to the triplet state  $T_1$  and are trapped if this decay occurs much faster than that of triplets back to the ground singlet states (i.e.  $\tau_{isc} \ll \tau_t$ ). The resulting reduction in the number of molecules in the singlet states leads to a reduced population inversion, which implies a lower gain. Moreover, a

high population of triplet states will also increase the lasing photon absorption and the number of molecules that are permanently lost through photobleaching. Molecules that make it to  $N_1$  then decay nonradiatively to  $N_0$  with a lifetime  $\tau_{10}$ . If the pumping rate exceeds the net decay rate of molecules in  $N_2$ , then a population inversion between  $N_2$  and  $N_1$  can be easily achieved because  $N_2$  depopulates slower compared to  $N_1$  and  $N_3$  (i.e.  $\tau_{21} \gg \tau_{10} \approx \tau_{32}$ ). Lasing occurs beyond a threshold  $P_{\text{src}}^{\text{thr}}$  when the inversion leads to a gain that is sufficiently large to overcome the total losses in the dye and the cavity. Dye absorptions of the emitted lasing photons also occur in the singlet states where the molecules in  $S_0$  and  $S_1$  are optically excited to  $S_1$  and  $S_2$  respectively.

### A. Subwavelength-structured cavity effects

We now turn to considering the subwavelength-structured cavity effects on the excited state emission lifetime of the molecules. The significant modification of the optical density of states in this class of structures with respect to the free-space case leads, in turn, to a substantial change of the corresponding spontaneous emission rate with respect to free-space emission. Specifically, within the Weisskopf-Wigner approximation, the spontaneous emission rate is proportional to the density of states<sup>28</sup>. We quantify this modification to the spontaneous emission rate centered near the lasing frequency  $f_l$  by including the factor  $F_p$  into the rate equations. We have defined  $F_p = \gamma_{\text{all modes}}^{\text{cav}} / \gamma_{\text{all modes}}^{\text{bulk}}$ , where  $\gamma_{\text{all modes}}^{\text{cav}}$  is the total emission rate in the presence of a cavity and  $\gamma_{\text{all modes}}^{\text{bulk}} (= 1/\tau_{\text{spont}})$  is the total emission rate of the bulk media in free space ( $\tau_{\text{spont}}$  is the corresponding spontaneous emission lifetime of the free-space medium). The inclusion of  $F_p$  in the model allows us to study the influence of the modified emission rate on the onset of lasing in novel nanostructured cavity designs.

Thus, the modified spontaneous emission rate in the presence of a cavity suitably designed for such a purpose can be expressed as<sup>29,30</sup>

$$\gamma_{f_l}^{\text{cav}} = \frac{\gamma_{f_l}^{\text{cav}}}{\gamma_{\text{all modes}}^{\text{cav}}} \times \frac{\gamma_{\text{all modes}}^{\text{cav}}}{\gamma_{\text{all modes}}^{\text{bulk}}} \times \gamma_{\text{all modes}}^{\text{bulk}} = \beta \times F_p \times \frac{1}{\tau_{\text{spont}}} \quad (1)$$

where  $\beta$  is the spontaneous emission coupling factor ( $\beta \equiv \gamma_{f_l}^{\text{cav}} / \gamma_{\text{all modes}}^{\text{cav}}$ ) and  $\gamma_{f_l}^{\text{cav}}$  is the emission rate into the lasing mode. In conventional, large cavities, where no enhancement of the spontaneous emission exists,  $F_p = 1$ .

## B. Continuous-wave lasing system

In this section, we focus on the operation of the considered lasing system under CW excitation. In order to do that, we analytically solve the coupled rate equations of Eq. (A1) to (A11) (Appendix A) in steady-state. The extension of these results to accommodate pulse excitations is presented in Sec. II C.

An analytical expression for the lasing threshold, that accounts for all relevant physical properties of the organic molecules and optical properties of the cavities, allows us to identify the relative importances of the various parameters in lowering the threshold under practical constraints. These constraints include the upper bound of a cavity's quality factor, the relative intersystem crossing lifetimes  $\tau_t/\tau_{isc}$  accessible to experiments, and the highest concentration of organic dye that one can use before its quantum yield reduces. Similarly, an analytical expression that predicts the photobleaching time of a dye will allow one to design systems that prolong the duration over which lasing action is possible. We separate the steady-state analysis into two parts: (i) predictions of the threshold and slope efficiency assuming no photobleaching, (ii) prediction of the time the dye takes to photobleach.

### 1. Threshold.

The relatively slow photobleaching process can be ignored in the predictions of thresholds since  $\tau_{bleach}$  is typically a few milliseconds (in air saturated solution at room temperature<sup>24,31</sup>) while the next longest lifetime is the triplet decay from  $T_1$  to  $S_0$  (i.e.  $\tau_t$ ), which varies between  $10^{-7}$  to  $10^{-4}$  seconds (in R6G depending if a solid-state or solution is used, or if a quencher is added). In other words, the effects of photobleaching only kick in after the lasing action begins. Under such assumption, the total molecular population is conserved in our model (i.e.  $dN_{tot}/dt = 0$ ) so that the gain will become saturable at a high pump rate. We limit our analysis to the regime near the threshold where the effects of the gain medium on the field remain linear<sup>32</sup>. We also assume that the total molecular population  $N_{tot} = N_{tot}^{den} \times V = N_0 + N_2 + N_4$  because molecules in the other levels are depleted due to their fast relaxation rates (this assumption breaks down only at very high excitation powers).  $N_{tot}^{den}$  is the density of dye molecules.

The steady state threshold power  $P_{src}^{thr}$  is then found by extrapolation of the linear input-output



power curve to zero output, which yields:

$$\frac{\eta^{S_0S_1} P_{\text{src}}^{\text{thr}}}{hf_p} = \frac{V (F_p/\tau_{\text{spont}} + 1/\tau_{21} + 1/\tau_{\text{isc}}) / \tau'_{\text{loss}}}{F_p \beta V \Gamma_s / \tau_{\text{spont}} - (1 + \tau_t/\tau_{\text{isc}}) / N_{\text{tot}}^{\text{den}} \tau''_{\text{loss}}} \quad (2)$$

where  $\eta^{S_0S_1}$  is the fraction of pump power absorbed by the  $S_0S_1$  transition of the organic medium,  $h$  is the Planck's constant, and  $f_p$  is the excitation frequency.  $1/\tau'_{\text{loss}} = 1/\tau_{\text{loss}} + v_g \sigma_{\text{abs}}^{S_0S_1} \Gamma_s N_{\text{tot}}^{\text{den}}$  and  $1/\tau''_{\text{loss}} = 1/\tau_{\text{loss}} + v_g \sigma_{\text{abs}}^{T_1T_2} \Gamma_s N_{\text{tot}}^{\text{den}}$  are the total loss-rates of the lasing photons; the first and second summands account for losses in the cavity and absorptions in the organic medium, respectively.  $\tau'_{\text{loss}}$  accounts for the absorptions in the singlet states while  $\tau''_{\text{loss}}$  accounts for those in the triplet states.  $\tau_{\text{loss}}$  is the photon decay lifetime in the passive cavity and is related to its total quality factor by  $Q_{\text{tot}} = 2\pi f_l \tau_{\text{loss}}$ . It accounts for the losses due to radiation into the far-field, scattering from surface roughness and intrinsic material absorption in actual devices.  $v_g$  is the group velocity of the lasing mode and  $\Gamma_s$  is its fractional energy contained in the organic region. In deriving Equation (2),  $\beta \Gamma_s$  is assumed to be much less than one and the relative magnitudes of the self-absorption cross sections are assumed to follow that in Table I of Appendix B for R6G in solution. Since  $\sigma_{\text{abs}}^{S_1S_2} \approx \sigma_{\text{abs}}^{T_1T_2}$ , the total photon loss rates in both the  $S_1S_2$  and  $T_1T_2$  states may be represented by  $1/\tau''_{\text{loss}}$  in our equations. We also note that the  $\beta V$  quantity is typically regarded to be volume-independent because  $\beta$  generally scales as the inverse of  $V$  in bulk systems<sup>33–35</sup>.

From the denominator of Equation (2), it becomes clear that lasing can only be realized in systems that satisfy

$$\frac{F_p \beta \Gamma_s N_{\text{tot}}}{\tau_{\text{spont}}} > \frac{(1 + \tau_t/\tau_{\text{isc}})}{\tau''_{\text{loss}}}. \quad (3)$$

The above relationship implies that in laser systems where the intrinsic losses are higher than the maximal gain available, lasing cannot be attained regardless of the pump power. To overcome this, one should thrive to increase the concentration of the organic media (while taking care to maintain high quantum yield), increase the confinement and  $\beta$  factors through optimizing the cavity designs, and reduce the losses in both dyes and cavities.

In particular, nanostructured materials, such as distributed feedback structures and photonic crystals, can be engineered to support delocalized resonances with long decay lifetimes, leading to enhanced interactions of light with the organic molecules<sup>12,36</sup>. Moreover, these nanostructures may increase the  $\beta$  factors through modification of the spectral density of states<sup>37</sup>, where both the spectral and angular radiation of the molecules are altered (when compared to their free-space emission)<sup>36</sup>. In view of Eq. (3), it is also important to realize that in the case of multimode laser systems featuring broad gain linewidths (so that a large number of cavity modes lie within the gain

curve), the Purcell enhancement is not expected to influence significantly the lasing threshold – essentially because in this case the Purcell increase on the magnitude of  $F_p$  is cancelled out by the corresponding decrease of the  $\beta$  factor.

## 2. Efficiency.

The quantum slope efficiency of the laser is defined as the probability that a lasing photon will be emitted with the absorption of a single pumping photon. It is proportional to the linear slope of the input-output power relationship above the threshold. Within our theoretical formalism, the quantum slope efficiency  $q_{\text{lase}}$  is given by:

$$q_{\text{lase}} = \frac{\Delta P_{\text{out}}}{\Delta P_{\text{src}}} \cdot \frac{f_p}{f_l} = \frac{\eta^{S_0 S_1}}{\tau_{\text{cav}}^{\text{rad}} \Gamma_s} \times \frac{F_p \beta V \Gamma_s / \tau_{\text{spont}} - (1 + \tau_t / \tau_{\text{isc}}) / N_{\text{tot}}^{\text{den}} \tau_{\text{loss}}''}{F_p \beta V / \tau_{\text{spont}} \tau_{\text{loss}} + \nu_g \sigma_{\text{abs}}^{S_0 S_1} [1 / \tau_{\text{loss}} + (1 + \tau_t / \tau_{\text{isc}}) / \tau_{\text{loss}}'']}. \quad (4)$$

$\tau_{\text{cav}}^{\text{rad}}$  is the photon decay lifetime of the cavity due to radiation into the far-field. The numerator in the second fraction of Eq. (4) is equivalent to the denominator of the threshold expression in Eq. (2), consistent with the fact that a low-threshold lasing system usually has high slope efficiency. In fact, if we replace the second fraction in Eq. (4) with the total photon-loss lifetime of the laser system  $\tau_{\text{loss}}^{\text{tot}}$ , a simple relationship for the slope efficiency can be found:  $\Delta P_{\text{out}} / \Delta P_{\text{src}} \propto \eta^{S_0 S_1} \tau_{\text{loss}}^{\text{tot}} / \tau_{\text{cav}}^{\text{rad}} \approx \eta^{S_0 S_1} P_{\text{cav}}^{\text{rad}} / P_{\text{loss}}^{\text{tot}}$ . In other words, the laser's slope efficiency depends on both the fraction of pump power that is absorbed by the organic medium and the fraction of the emitted power that is radiated into the far-field.

## 3. Photobleaching.

Next, we include the photobleaching channel in  $N_4$  through which a molecule can become permanently lost and not return to the singlet states to further participate in the stimulated emission<sup>23</sup>. In this case, the total molecular population is no longer conserved but reduces according to  $dN_{\text{tot}}/dt = -N_4/\tau_{\text{bleach}}$ , where  $\tau_{\text{bleach}}$  can be markedly different depending on the environment of the dyes (see Appendix A). This sets the time scale  $\tau_{\text{system}}^{\text{bleach}} = \tau_{\text{bleach}} N_{\text{tot}} / N_4$  beyond which the dye photobleaches and the emitted signal is permanently quenched. In deriving Eq. (5) below, the steady-state populations of  $N_4$  and  $N_{\text{tot}}$  (assuming no photobleaching as in Sec. II B 1 and II B 2) are substituted into the expression for  $\tau_{\text{system}}^{\text{bleach}}$  above, even though  $N_4$  and  $N_{\text{tot}}$  are now time-dependent quantities. We checked the validity of such an approximation by comparing the numerically pre-

dicted photobleaching rates using the full rate equations with the analytically predicted ones, and found their comparison to be good (illustrated in Sec. III). As a passing remark, we point out that our model predicts  $\tau_{\text{system}}^{\text{bleach}}$  to decrease linearly with the excitation power when operating below the lasing threshold. This linear trend of the bleaching rate with pump power is consistent with experimental observations<sup>13</sup>.

The photobleaching lifetime upon lasing (a volume-independent quantity) can be expressed as<sup>49</sup>

$$\begin{aligned}\tau_{\text{lase}}^{\text{bleach}} &= \tau_{\text{bleach}} \times F_{\text{lase}}^{\text{bleach}} \\ &\approx \tau_{\text{bleach}} \times \frac{N_{\text{tot}}^{\text{den}} \Gamma_s \tau'_{\text{loss}}}{\tau_t / \tau_{\text{isc}}} \cdot \left[ \frac{F_p \beta V}{\tau_{\text{spont}}} - v_g \sigma_{\text{abs}}^{T_1 T_2} \left( 1 + \frac{\tau_t}{\tau_{\text{isc}}} \right) \right]\end{aligned}\quad (5)$$

Beyond this lifetime, the dye photobleaches and lasing shuts down.  $F_{\text{lase}}^{\text{bleach}}$  is the modification to  $\tau_{\text{bleach}}$  afforded by the specific design of the laser system; it depends on the dye concentration and intersystem crossing lifetime ratio ( $\tau_t / \tau_{\text{isc}}$ ), as well as the cavity's quality factor, confinement factor, spontaneous emission coupling factor ( $\beta$ ) and spontaneous emission modification factor ( $F_p$ ). Again, the relative values of the absorption cross-sections in Table I are assumed in deriving Eq. (5).

The quantum efficiency for dye bleaching, which gives the probability that a dye molecule will be bleached with the absorption of a single pumping photon, may similarly be derived as

$$q_{\text{bleach}} \approx \frac{N_{\text{tot}} h f_p}{\tau_{\text{system}}^{\text{bleach}} \eta_{S_0 S_1} P_{\text{src}}}. \quad (6)$$

$q_{\text{bleach}}$  is independent of the organic concentration. Its reciprocal  $1/q_{\text{bleach}}$  provides an estimate of the number of times a molecule is recycled from  $S_0$  to  $S_1$  and back to  $S_0$  before it is lost. Since  $\tau_{\text{system}}^{\text{bleach}} \propto 1/P_{\text{src}}$  as noted above,  $q_{\text{bleach}}$  in Eq. (6) is also independent of the excitation power (below the threshold).

### C. Pulsed lasing system

Most lasing experiments involving organic gain materials are excited by short pulses<sup>8,9,38</sup>, except for systems where liquid dye circulation exist<sup>14,22</sup>. The key difference of the analysis in this section from that of the steady-state is that the undesirable triplet influences can now be minimized when the excitation pulse-width  $\tau_{\text{pulse}}$  is shorter than the intersystem crossing lifetime  $\tau_{\text{isc}}$ . This decoupling between the singlet and triplet states can be represented in our analytic model

by setting  $\tau_{\text{isc}}$  to large values so that  $N_{\text{tot}}^{\text{pulse}} = N_0 + N_2$  at all times and no photobleaching takes place within the short lifespan of the lasing pulse signal. Physically, a large  $\tau_{\text{isc}}$  implies a quasi steady-state regime where the pulse length is assumed to be longer than all decay lifetimes of the molecules but remains shorter than  $\tau_{\text{isc}}$ .

Moreover, the reabsorption of the lasing photons in  $S_1S_2$  is sometimes neglected because the population of  $S_0$  is much greater than that of  $S_1$  so that  $N_0\sigma_{\text{abs}}^{S_0S_1}$  is the dominant photon loss term in Eq. (A11) of Appendix A. As a result, a four-level gain model of the lower singlet states usually suffices in capturing the main lasing mechanisms. In this work, however, we include the photon absorption in  $S_1S_2$  (since  $\sigma_{\text{abs}}^{S_1S_2} \gg \sigma_{\text{abs}}^{S_0S_1}$  in some dyes such as R6G) and show that it plays a non-negligible role on the threshold values when  $1/\tau_{\text{loss}}$  in  $1/\tau_{\text{loss}}''$  becomes relatively small (i.e.  $1/\tau_{\text{loss}}'' \approx \nu_g \sigma_{\text{abs}}^{S_1S_2} \Gamma_s N_{\text{tot}}/V$ ). Using the parameters of our lasing experiment<sup>36</sup>, this happens when  $Q_{\text{tot}} = 2\pi f_l \tau_{\text{loss}} > 5 \times 10^5$  which is a physically realizable value<sup>39</sup>.

By setting  $\tau_{\text{isc}}$  to infinity in Eq. (2), the threshold pulse energy in the quasi steady-state regime is approximated as

$$U_{\text{pulse}}^{\text{thr}} = P_{\text{src}}^{\text{thr}} \tau_{\text{pulse}} \approx \frac{hf_p \tau_{\text{pulse}}}{\eta^{S_0S_1}} \times \frac{(F_p/\tau_{\text{spont}} + 1/\tau_{21})/\tau_{\text{loss}}'}{F_p \beta V \Gamma_s / \tau_{\text{spont}} - 1/N_{\text{tot}}^{\text{den}} \tau_{\text{loss}}''} \times V. \quad (7)$$

In most pulsed lasing systems,  $\tau_{\text{pulse}}$  and  $\tau_{\text{spont}}$  are both on the order of a few nanoseconds and thus, the quasi steady-state approximation applied in Eq. (7) leads to an underestimation of the threshold pulse energy. Nonetheless, Eq. (7) is still useful in identifying the key molecular and photonic parameters needed to lower the lasing threshold. Similar to the steady-state regime of Eq. (3), lasing can only take place when the maximal net gain is greater than the total cavity and molecular losses where  $F_p \beta \Gamma_s N_{\text{tot}}/\tau_{\text{spont}} > 1/\tau_{\text{loss}}''$ . However, unlike in Eq. (3), these losses are no longer influenced by the triplet states.

### 1. Comparison with experiment.

To verify our model, a pulsed lasing experiment with  $1mM$  R6G dissolved in methanol on top of a photonic crystal slab was conducted<sup>36</sup>, and its lasing threshold and slope are compared to numerical predictions. Fabrication and characterization of the passive photonic crystal cavity has been reported in a previous study by Lee *et al.*<sup>40</sup>. The active cavity is optically excited near the peak of the dye's absorption spectrum at  $\lambda_p = 532 \text{ nm}$  with  $5 \text{ ns}$  pulses at a repetition rate

of 10 Hz. Lasing occurs at  $\lambda_{\text{lase}} = 580 \text{ nm}$  where a strong optical resonance exists well within the R6G's emission spectrum, and having a confinement factor  $\Gamma_s$  of 7%.  $Q_{\text{tot}}$  is measured to be  $8.3 \times 10^3$ ,  $Q_{\text{cav}}^{\text{rad}}$  is calculated to be  $1.9 \times 10^5$ , while  $\beta$  is approximated as  $7 \times 10^{-5}$  using the expression for bulk systems discussed in Appendix A. We input these parameters into our lasing model and observe that both the threshold and slope match well with the measured data, within experimental errors<sup>36</sup>. In particular, the measured threshold is 100 nJ while that predicted from our lasing model is 75 nJ.

### III. RESULTS AND DISCUSSION

In this section, we analyze the steady-state operation of the R6G organic laser in terms of its threshold and the duration over which lasing action can be sustained before the dye photobleaches. The optical and physical properties of the system in Eq. (2) and (5) are varied and their influences on the lasing performances are studied. The parameter values used in our examples are listed in Table I of Appendix B for R6G in solution. We begin the analysis by verifying the steady-state analytic expressions derived in Sec. II. In particular, the analytic solutions are compared to the numerical solutions of the full model.

Figure 2(a) illustrates the output intensity  $I_{\text{out}}$  (red curve) and photobleaching lifetime (blue curve) of the laser as a function of the excitation intensity  $I_{\text{src}}$ .  $I_{\text{out}}$  plotted corresponds to the quasi steady-state values before the laser emission is quenched [see Figure 2(b)]. In this example,  $\tau_t$  is assumed to be 10  $\mu\text{s}$  so that  $\tau_t/\tau_{\text{isc}} = 100$  and the threshold is found to be near 1  $\text{W}/\text{cm}^2$ . By plotting the results against  $\eta I_{\text{src}}$  in Fig. 2(a), we are able to account for the portion of incident power that is not absorbed by the dye; when  $\eta < 1$ , the threshold intensity would have to be scaled by  $1/\eta$ . For instance, if only 10% of the incident power is absorbed by the dye laser, then, the threshold becomes 10  $\text{W}/\text{cm}^2$  instead. We also found that in this example, the quantum slope efficiency of the laser (calculated from the linear input-output intensity plot beyond threshold) is roughly 40% when  $\eta = 1$ . This matches the prediction of Eq. (4) and is also consistent with the simple relationship of  $q_{\text{lase}} = \Delta P_{\text{out}} f_p / \Delta P_{\text{src}} f_l \approx \eta P_{\text{cav}}^{\text{rad}} / P_{\text{loss}}^{\text{tot}}$  presented in Sec. II. Specifically,  $P_{\text{cav}}^{\text{rad}} / P_{\text{loss}}^{\text{tot}} = 50\%$  for the passive cavity considered in Table I and further reduction to 40% arises when the absorption losses in the dye's triplet states are included.

Next, we examine the photobleaching lifetime of the dye system under CW excitation. From Fig. 2(a),  $\tau_{\text{system}}^{\text{bleach}}$  decreases linearly with the excitation intensity below threshold and clamps at its

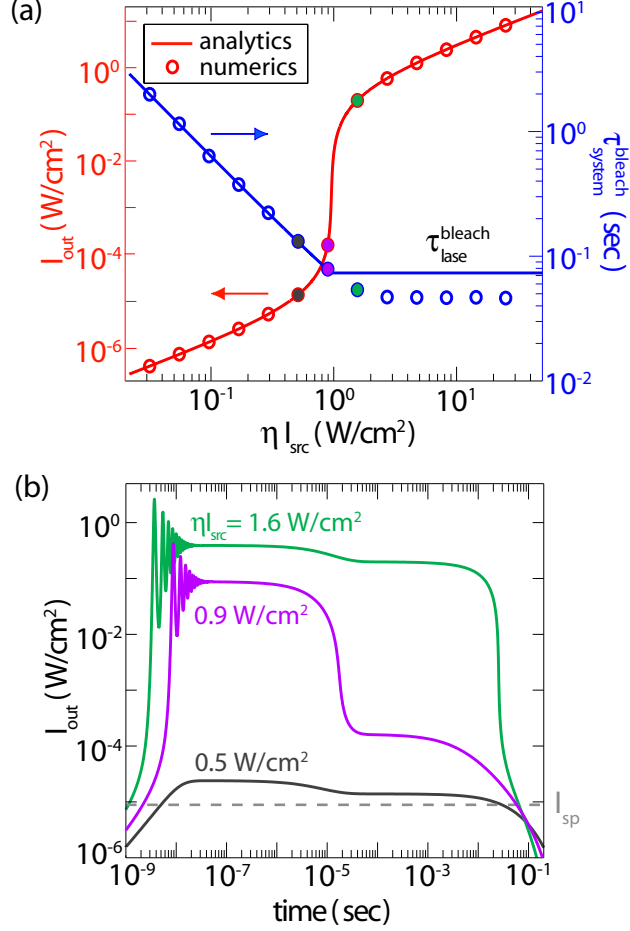


FIG. 2: (a) Output intensity  $I_{\text{out}}$  (red curve) and photobleaching lifetime (blue curve) of the organic laser as a function of the CW excitation intensity  $I_{\text{src}}$ . The plotted  $I_{\text{out}}$  correspond to the quasi steady-state values before the laser emission is quenched [see plot in (b)]. The circles are results obtained from numerically solving the full set of rate equations in Appendix A while the solid lines are steady-state analytical solutions (outlined in Sec. II B) to the same rate equations. The filled circles correspond to the three excitation intensities analyzed in (b).  $\eta$  is the fraction of the pump power absorbed by the gain medium. (b) Temporal development of lasing action of the laser system in (a) for three  $\eta I_{\text{src}}$  values near threshold.  $I_{\text{sp}}$  is the intensity level of spontaneous emission (see text for details on its definition).

threshold value upon lasing. To understand this behavior, we first compute the bleaching quantum efficiency in Eq. (6) and find  $q_{\text{bleach}} = 5 \times 10^{-5}$ . This implies that every molecule in the dye laser can be recycled to produce roughly  $q_{\text{lase}}/q_{\text{bleach}} = 8 \times 10^3$  emitted photons before it is annihilated in  $N_4$ . When the pump rate is high, the molecules are recycled through the  $S_0S_1$  states at a faster rate; each molecule reaches its maximum emission of  $8 \times 10^3$  photons in a shorter time period.

Hence, the photobleaching rate increases linearly with the excitation intensity. However, once the lasing is attained, the additional pumping power above threshold is channeled entirely into the coherently oscillating cavity mode and  $\tau_{\text{system}}^{\text{bleach}}$  clamps at its threshold value of  $\tau_{\text{lase}}^{\text{bleach}}$  [Eq. (5)].

We note that  $q_{\text{bleach}}$  can be reduced by up to two orders of magnitude if an appropriate triplet quencher for R6G is used to reduce  $\tau_t/\tau_{\text{isc}}$  from 100 to 1. It can also be reduced by lengthening  $\tau^{\text{bleach}}$  through chemically modifying the environment of the R6G molecules. For example, it is known that lowering water and oxygen results in a decreased photobleaching rate. Care should, however, be taken in this case to manage the parallel increase in triplet state population (oxygen is also a quencher of the triplet states)<sup>25</sup> to ensure a net reduction of  $q_{\text{bleach}}$ .

As seen in Fig. 2(a), the agreement between the analytical and numerical results are excellent, except for the  $\tau_{\text{lase}}^{\text{bleach}}$  value which differs by a factor of  $\sim 1.5$ . The mismatch occurs because the analytical solutions consider the steady-state  $N_4$  and  $N_{\text{tot}}$  values (without photobleaching) even though they are in fact time-dependent quantities (see Sec. II). Nonetheless, this discrepancy remains smaller than a factor of 2 in all the examples considered and does not affect the conclusions drawn.

Figure 2(b) shows the temporal development of lasing action in the same laser for three  $\eta I_{\text{src}}$  values near threshold. Below threshold at  $\eta I_{\text{src}} = 0.5 \text{ W/cm}^2$ , the output intensity is close to the spontaneous emission level  $I_{\text{sp}}$ . Within the semiclassical picture of spontaneous emission, this intensity corresponds to the energy of one photon<sup>41</sup> so that  $I_{\text{sp}} = hf_l/\tau_{\text{cav}}^{\text{rad}}A_p$ .  $A_p$  is the area under illumination. When  $\eta I_{\text{src}}$  is increased to  $0.9 \text{ W/cm}^2$ , the laser is turned on initially between  $10^{-8}\text{s}$  to  $10^{-5}\text{s}$  where the output intensity reaches more than three orders of magnitude above  $I_{\text{sp}}$ . However, the laser shuts down at  $10^{-5}\text{s}$  and the output decreases by two orders of magnitude to a quasi steady-state level before the dye photobleaches. The initial turn-on occurs when  $I_{\text{src}}$  is high enough to overcome the total losses in the singlet states. As the triplet states begin to populate, the associated absorption losses and trapping of molecules eventually quench the singlets inversion. At higher pumping rate of  $\eta I_{\text{src}} = 1.6 \text{ W/cm}^2$ , the laser is turned on and the stimulated emission of light is sustained even after the triplet excited state absorption kicks in. In all the three cases above, dye emission ceased after  $10^{-1}\text{s}$  due to photobleaching. Moreover, a series of very narrow spikes whose amplitudes die down with time can be observed upon turn on of the laser. These phenomena of spiking and relaxation oscillations are characteristics of laser systems in which the recovery time of the excited state population inversion is longer than the total decay time of the lasing mode<sup>41</sup>.

In Fig. 3 and Fig. 4, five parameters of the laser system are varied and their effects on the threshold ( $\eta I_{\text{src}}^{\text{thr}}$ ) and corresponding photobleaching modification factor ( $F_{\text{lase}}^{\text{bleach}}$ ) are studied. These results are plotted using Eq. (2) and (5) respectively. For illustration, the five parameters selected in this study are the total quality factor of the passive cavity ( $Q_{\text{tot}}$ ), intersystem crossing lifetime ratio ( $\tau_t/\tau_{\text{isc}}$ ), dye concentration, confinement factor ( $\Gamma_s$ ), and the spontaneous emission modification factor ( $F_p$ ). Practical values of  $Q_{\text{tot}}$  range from few hundreds to tens of millions<sup>39</sup> and we expect the threshold intensity to decrease with increasing  $Q_{\text{tot}}$ . A reduced pumping intensity for lasing will in turn lengthen the time it takes for the dye to photobleach. In the following, we varied  $Q_{\text{tot}}$  with each of the other four parameters above to understand their effects on the laser characteristics.

#### A. Intersystem crossing lifetime ratio ( $\tau_t/\tau_{\text{isc}}$ ) [Fig. 3(a) and 4(a)]

In Eq. (2) to (5), the ratio  $\tau_t/\tau_{\text{isc}}$  plays an important role in influencing the lasing performances. A high  $\tau_t/\tau_{\text{isc}}$  will not only increase the lasing threshold but also decrease the slope efficiency and lasing lifetime  $\tau_{\text{lase}}^{\text{bleach}}$ . These observations can be understood from the accompanying build-up in the triplet state population,  $N_4 = \tau_t/\tau_{\text{isc}} \times N_2$ , through which high molecular- and photon-losses occur. Hence, it is desirable that the organic gain media have small values of  $\tau_t/\tau_{\text{isc}}$ . In R6G, triplet quenchers which affect  $\tau_t$  are readily available and by simultaneously changing the environment of the molecules from solid to solution,  $\tau_t/\tau_{\text{isc}}$  can be varied over four orders of magnitudes from 1 to  $10^4$ . The threshold plot illustrates a cut-off beyond which lasing shut down as  $\tau_t/\tau_{\text{isc}}$  increases. This happens when the triplets influence is so high that its associated photon absorption and molecular annihilation process overwhelmed the stimulated emission of light. In other words, Eq. (3) cannot be satisfied. Note that high values of the lifetime ratio beyond  $10^3$  are typical in solid-state organic materials and based on our results, are major impediments to the attainment of low-threshold CW organic solid-state lasers. On the other hand, as  $\tau_t/\tau_{\text{isc}}$  decreases, the triplet states become less populated and the threshold values saturated towards that of a short-pulse system where only the energy transfers in the singlet states are relevant (see Sec. II C).

Unlike the threshold values, the photobleaching factor in Fig. 4(a) does not saturate but continue to increase with declining  $\tau_t/\tau_{\text{isc}}$ . This trend is directly related to the corresponding reduced population of the  $N_4$  state, where loss of the molecules takes place in our model. In our example, a threshold of  $\eta I_{\text{src}}^{\text{thr}} = 0.7 \text{ W/cm}^2$  and photobleaching factor of  $F_{\text{lase}}^{\text{bleach}} = 1125$  is predicted for the considered CW R6G organic laser when  $Q_{\text{tot}} = 5 \times 10^4$  and  $\tau_t/\tau_{\text{isc}} = 1$ .



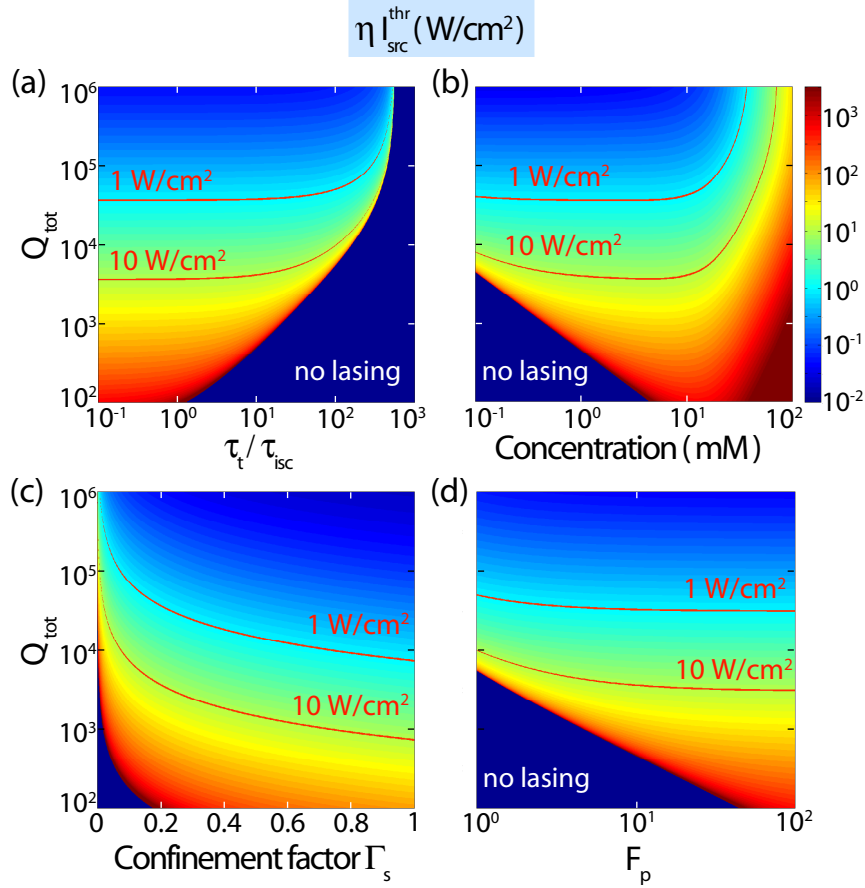


FIG. 3: Lasing threshold ( $\eta I_{src}^{thr}$ ) as a function of the optical and physical properties in a R6G organic laser under continuous excitation.  $\eta$  is the fraction of the excitation power absorbed by the dye. The parameter values used are listed in Table I, except for the plot in (d) where  $\tau_t$  is assumed to be  $10 \mu s$  so that  $\tau_t / \tau_{isc} = 100$  instead of 1. In particular, the total quality factor of the passive cavity ( $Q_{tot}$ ) is varied with the intersystem crossing lifetime ratio ( $\tau_t / \tau_{isc}$ ) in (a), the dye concentration in (b), the confinement factor ( $\Gamma_s$ ) in (c), and the spontaneous emission modification factor ( $F_p$ ) in (d).

### B. Dye concentration [Fig. 3(b) and 4(b)]

In diluted R6G with concentration below  $5 \text{ mM}$ , the quantum yield is high (i.e.  $q_{dye} = 0.9$ ) and  $\tau_{spont}$  is approximately  $5 \text{ ns}$ . As the concentration increases beyond  $10 \text{ mM}$ , both the quantum yield of the dye and its spontaneous emission lifetime begin to decrease<sup>42</sup>. We incorporate such concentration-dependent decrease in quantum yield and spontaneous emission lifetime into our model. From the threshold plot, we found that a minimum dye concentration is required for lasing to take place [i.e. Eq. (3) is satisfied] when the cavity losses are high; dilute organic so-

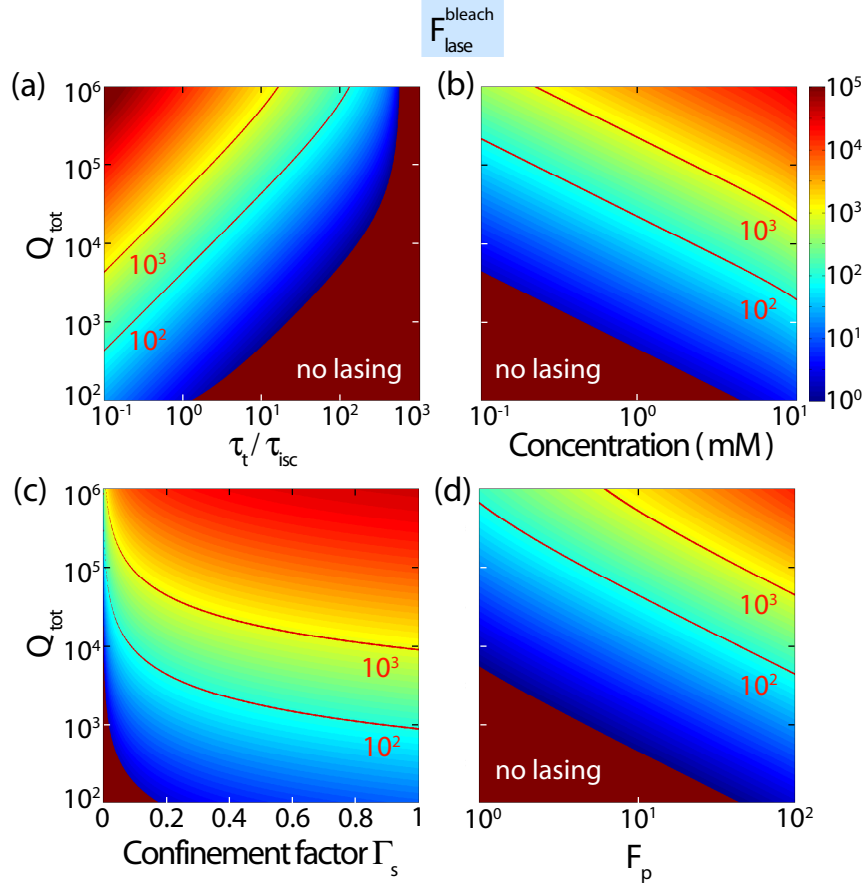


FIG. 4: Photobleaching modification factor ( $F_{\text{lase}}^{\text{bleach}}$ ) as a function of the optical and physical properties in the same R6G organic laser system of Fig. 3.

lutions cannot provide sufficient gain to overcome the total losses of the system. On the other hand, concentrated R6G solution above  $10 \text{ mM}$  is also undesirable because  $q_{\text{dye}}$  begins to decrease causing the threshold to rise rapidly. The decrease of  $q_{\text{dye}}$  is attributed to Förster-type excitation transfer from monomers to weakly fluorescing dimers, which are formed randomly at high concentrations<sup>42</sup>. Physically, a low  $q_{\text{dye}}$  implies only a small fraction of the molecules excited into  $N_2$  contributes to the emission of light. This leaves  $(5 - 10) \text{ mM}$  to be the optimal concentration to operate the laser with the lowest threshold.

In Fig. 4(b), the photobleaching factor is observed to increase with concentration (up to  $10 \text{ mM}$ ) for all values of  $Q_{\text{tot}}$ . This follows directly from the fact that a highly concentrated solution consists of more molecules (assuming the same volume of dye) able to participate in the excitation and de-excitation processes, and be recycled through the  $S_0S_1$  states before they become quenched from photobleaching.

### C. Confinement factor ( $\Gamma_s$ ) [Fig. 3(c) and 4(c)]

A higher confinement factor leads to a larger modal gain in the system for the same excitation level, resulting in a reduction of the excitation threshold intensity<sup>43</sup>. This reduction in the threshold in turn increases the photobleaching factor, as observed in Fig. 4(c), and equivalently slows down the photobleaching rate. However, the decrease in both the threshold and photobleaching rate are dampened by the enhanced self-absorption of the dye at higher  $\Gamma_s$ , and hence, only a marginal improvement of the laser performances with increasing  $\Gamma_s$  is observed. The above description is applicable for  $\Gamma_s > 0.1$  for the range of  $Q_{\text{tot}}$  considered. When  $\Gamma_s < 0.1$ , the laser performances deteriorate rapidly.

### D. Spontaneous emission modification factor ( $F_p$ ) [Fig. 3(d) and 4(d)]

In this example,  $\tau_t$  is assumed to be  $10 \mu\text{s}$  so that  $\tau_t/\tau_{\text{isc}} = 100$  instead of 1 as considered in Table I.  $F_p$  is the modification to the spontaneous emission rate of the free-space bulk organic dye ( $1/\tau_{\text{spont}}$ ) in the presence of a cavity. It can be high in microcavities ( $F_p = 75$ ) with small mode volumes, while maintaining its influence over a reasonably large spectrum of the emitter's bandwidth (quality factor of  $\sim 80$ )<sup>44</sup>. From the threshold plot, we observed that  $F_p$  plays an important role in enabling lasing, especially at low  $Q_{\text{tot}}$ . However, its impact on threshold reduction diminishes as  $F_p$  increases; the  $F_p$  factors in the numerator and denominator of Eq. (2) cancel out when the leading terms become dominating at large values of  $F_p$ . The physical interpretation is that a high  $F_p$  will not only lead to a desirable enhancement in the stimulated emission of light but also increase the spontaneous emission rate. Since the spontaneously emitted light is characterized as noise in a laser, the latter process is undesirable once lasing has taken place. In particular, spontaneous emission becomes a competing radiative process to stimulated emission through which molecules can decay radiatively out of the  $N_2$  metastable state. When  $F_p$  becomes large, these two processes (stimulated and spontaneous emission) become equally dominant and the resulting reduction in threshold saturates.

On the other hand, the photobleaching lifetime of the dye does not saturate but continue to lengthen as  $F_p$  increases. This trend is directly related to the reduced population of the  $N_4$  state when the total stimulated and spontaneous emission rates are enhanced.

In the analysis above, we have kept  $\beta$  unchanged to isolate the influence of  $F_p$ . We emphasize

that, as mentioned in Sec. II B 1, in multimode cavities, when the gain linewidth is much broader than the cavity mode spacing, the reduction of the lasing threshold achieved by a larger value of  $F_p$  will be negated by the corresponding decrease of  $\beta$ .

#### IV. CONCLUSIONS

We have presented a comprehensive theoretical analysis of lasing action in systems of subwavelength-structured cavities consisting of organic dye molecules. The formalism considers both the physical properties of the dyes and optical properties of the cavities, and provides explicit analytic expressions of the threshold and slope efficiency that characterize this class of lasers. Quenching of the laser emission due to photobleaching of the dyes is also considered. To illustrate this theoretical framework, we have analyzed in detail the case of lasing action from optically pumped monolithic R6G organic dye lasers. Our simulations have shown that for this system, with exemplary parameters listed in table I, a value for the threshold  $\eta I_{\text{src}}^{\text{thr}}$  of a few  $W/cm^2$  can be achieved. Nanostructured materials that possess high quality factors and high spontaneous emission factors have been demonstrated<sup>36,45</sup> and are ideal resonators for realizing low-threshold organic lasers<sup>12</sup>. We believe our results could contribute to enabling the advancement of nanostructured organic light emitting and sensing devices, as well as expanding the current understanding of gain-compensation and lasing action in plasmonic and metamaterial-based active devices.

#### V. ACKNOWLEDGMENTS

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## APPENDIX A: RATE EQUATIONS

In this appendix, we first present the set of the spatially-averaged coupled rate equations that are used to model the organic gain medium described by the processes shown in Fig. 1. As a general energy-level model of an organic molecule, we consider the three lowest singlet and two lowest triplet states. We then relate the optically-induced transition terms of our model to the experimentally-measured cross-sections  $\sigma$ , and show how such formalism can incorporate the influences of the cavity through the spontaneous emission modification factor  $F_p$  and spontaneous emission coupling factor  $\beta$ .

The rate equations of the molecular populations in each level ( $N_0$  to  $N_9$ ), and the semiclassical rate equation describing the lasing photon number  $\phi_s$  can be written as

$$\frac{dN_0}{dt} = -\frac{P_{\text{src}}}{hf_p} \eta^{S_0S_1} \frac{(N_0 - N_3)}{N_{\text{tot}}} - \nu_g \sigma_{\text{abs}}^{S_0S_1} \Gamma_s \phi_s \frac{(N_0 - N_2)}{V} + \frac{N_1}{\tau_{10}} + F_p \frac{(1 - \beta \Gamma_s)}{\tau_{\text{spont}}} N_2 \quad (\text{A1})$$

$$\frac{dN_1}{dt} = +F_p \frac{\beta}{\tau_{\text{spont}}} \Gamma_s \phi_s (N_2 - N_1) + N_2 \left( F_p \frac{\beta \Gamma_s}{\tau_{\text{spont}}} + \frac{1}{\tau_{21}} \right) + \frac{N_4}{\tau_t} - \frac{N_1}{\tau_{10}} \quad (\text{A2})$$

$$\begin{aligned} \frac{dN_2}{dt} = & -F_p \frac{\beta}{\tau_{\text{spont}}} \Gamma_s \phi_s (N_2 - N_1) - \nu_g \sigma_{\text{abs}}^{S_1S_2} \Gamma_s \phi_s \frac{(N_2 - N_8)}{V} - N_2 \left( \frac{F_p}{\tau_{\text{spont}}} + \frac{1}{\tau_{21}} + \frac{1}{\tau_{\text{isc}}} \right) \\ & - \frac{P_{\text{src}}}{hf_p} \eta^{S_1S_2} \frac{(N_2 - N_9)}{N_{\text{tot}}} + \nu_g \sigma_{\text{abs}}^{S_0S_1} \Gamma_s \phi_s \frac{(N_0 - N_2)}{V} + \frac{N_3}{\tau_{32}} \end{aligned} \quad (\text{A3})$$

$$\frac{dN_3}{dt} = +\frac{P_{\text{src}}}{hf_p} \eta^{S_0S_1} \frac{(N_0 - N_3)}{N_{\text{tot}}} + \frac{N_8}{\tau_{83}} - \frac{N_3}{\tau_{32}} \quad (\text{A4})$$

$$\frac{dN_4}{dt} = -\frac{P_{\text{src}}}{hf_p} \eta^{T_1T_2} \frac{(N_4 - N_7)}{N_{\text{tot}}} - \nu_g \sigma_{\text{abs}}^{T_1T_2} \Gamma_s \phi_s \frac{(N_4 - N_6)}{V} + \frac{N_2}{\tau_{\text{isc}}} + \frac{N_5}{\tau_{54}} - \frac{N_4}{\tau_t} - \frac{N_4}{\tau_{\text{bleach}}} \quad (\text{A5})$$

$$\frac{dN_5}{dt} = +\frac{N_6}{\tau_{65}} - \frac{N_5}{\tau_{54}} \quad (\text{A6})$$

$$\frac{dN_6}{dt} = +\nu_g \sigma_{\text{abs}}^{T_1T_2} \Gamma_s \phi_s \frac{(N_4 - N_6)}{V} + \frac{N_7}{\tau_{76}} - \frac{N_6}{\tau_{65}} \quad (\text{A7})$$

$$\frac{dN_7}{dt} = +\frac{P_{\text{src}}}{hf_p} \eta^{T_1T_2} \frac{(N_4 - N_7)}{N_{\text{tot}}} - \frac{N_7}{\tau_{76}} \quad (\text{A8})$$

$$\frac{dN_8}{dt} = +\nu_g \sigma_{\text{abs}}^{S_1S_2} \Gamma_s \phi_s \frac{(N_2 - N_8)}{V} + \frac{N_9}{\tau_{98}} - \frac{N_8}{\tau_{83}} \quad (\text{A9})$$

$$\frac{dN_9}{dt} = +\frac{P_{\text{src}}}{hf_p} \eta^{S_1S_2} \frac{(N_2 - N_9)}{N_{\text{tot}}} - \frac{N_9}{\tau_{98}} \quad (\text{A10})$$

$$\begin{aligned} \frac{d\phi_s}{dt} = & -\frac{\nu_g \Gamma_s \phi_s}{V} \left[ \sigma_{\text{abs}}^{S_0S_1} (N_0 - N_2) + \sigma_{\text{abs}}^{S_1S_2} (N_2 - N_8) + \sigma_{\text{abs}}^{T_1T_2} (N_4 - N_6) \right] - \frac{\phi_s}{\tau_{\text{loss}}} \\ & + F_p \frac{\beta}{\tau_{\text{spont}}} \Gamma_s \phi_s (N_2 - N_1) + F_p \frac{\beta}{\tau_{\text{spont}}} \Gamma_s N_2. \end{aligned} \quad (\text{A11})$$

It is implicitly implied in the above that  $P_{\text{src}}$ ,  $N_i$  ( $i = 0$  to  $9$ ), and  $\phi_s$  are time-dependent quantities.  $\eta^{S_0S_1}$ ,  $\eta^{S_1S_2}$  and  $\eta^{T_1T_2}$  are the fractions of pump power absorbed by the  $S_0S_1$ -,  $S_1S_2$ - and  $T_1T_2$ -transition of the organic dye, respectively.  $N_{\text{tot}} = \sum_{i=0}^9 N_i = N_{\text{tot}}^{\text{den}} \times V$  is the total molecular population obtained by taking the product of the molecular density and volume of the organic medium, and  $\tau_{ij}$  is the non-radiative decay lifetime of the molecules from level  $i$  to  $j$ . In particular,  $\tau_{21}$  is the non-radiative lifetime in  $N_2$  and is related to the spontaneous emission lifetime by the dye's quantum yield  $q_{\text{dye}}$ , where  $\tau_{21} = \tau_{\text{spont}} \times q_{\text{dye}} / (1 - q_{\text{dye}})$ . Within our theoretical formalism, the laser output is computed as  $P_{\text{out}} = hf_l \phi_s / \tau_{\text{cav}}^{\text{rad}}$ , where  $\tau_{\text{cav}}^{\text{rad}}$  is the photon decay lifetime of the passive cavity due to radiation into the far-field.

*Excitation of molecules.* In the excitation terms of the rate equations,  $P_{\text{src}}$  is the source power and  $\eta^{S_0S_1}$  is the fraction of pump power absorbed by the  $S_0S_1$  transition of the organic medium. The pump terms excite molecules from the  $S_0$ ,  $S_1$  and  $T_1$  states to their respective higher energy levels separated by the photon energy  $hf_p$ . In a single-pass absorption of the pump light over a distance  $l$ ,  $\eta^{S_0S_1}$  is typically approximated by  $\sigma_{\text{pump}}^{S_0S_1} \Gamma_p l N_{\text{tot}}^{\text{den}}$  where  $\sigma_{\text{pump}}^{S_0S_1}$  is the experimentally measured absorption cross section of the bulk medium at the excitation wavelength and  $\Gamma_p$  is the fractional energy of the pumping mode contained in the organic medium. However, in cases where the pump light is resonantly coupled into the organic medium and the absorption of light occurs over multiple passes, the relationship between  $\eta$  and  $\sigma_{\text{pump}}$  is less trivial. For instance,  $\eta^{S_0S_1}$  is estimated by considering the total losses of the system (both absorption in the dye as well as radiation and scattering losses of the cavity) and applying the  $Q$ -matching arguments<sup>46</sup> when we compared the experimentally measured threshold and slope efficiency values to numerical predictions<sup>36</sup>.

Our numerical simulations of the full model considered pumping transitions in  $S_0S_1$ ,  $S_1S_2$  and  $T_1T_2$  but only the excitation of the molecules from  $S_0$  to  $S_1$  is significant because its absorption cross section is an order of magnitude larger than those in  $S_1S_2$  and  $T_1T_2$ . Moreover, the population of the ground singlet state is also larger than those in  $S_1$  and  $T_1$ , which further diminishes the importance of both  $S_1S_2$  and  $T_1T_2$  pumping transitions when operating at power levels near the lasing threshold. Thus, in deriving the analytic solutions in Sec. II of the main text, only the pumping transition from  $S_0$  to  $S_1$  is considered. Apart from the pump-induced excitations of the molecules, the emitted lasing light is also reabsorbed by the dye to excite molecules from the  $S_0$ ,  $S_1$  and  $T_1$  states to their respective higher energy levels separated by the photon energy  $hf_l$ . These self-absorption transitions are represented in the rate equations using the cross sections  $\sigma_{\text{abs}}$ .

*Cavity effects.* We now explain how the influences of the cavity are introduced into the stimulated and spontaneous emission terms of the rate equations. We begin by relating the experimentally measured stimulated emission cross sections  $\sigma_{se}$  to the spontaneous emission coupling factor  $\beta$ .  $\sigma_{se} = \lambda_l^4 g(\lambda_l) / 8\pi c n_{eff}^2 \tau_{spont}$  in bulk organic media<sup>42</sup> and  $\beta = \lambda_l^4 g(\lambda_l) / 8\pi n_{eff}^3 V$  in bulk systems<sup>33-35</sup> (i.e. large resonators). In both relationships,  $g(\lambda_l)$  is the photoluminescence spectrum normalized to the fluorescence quantum yield  $q_{dye}$ <sup>42</sup>,  $n_{eff}$  is the effective index of the lasing mode, and  $c$  is the speed of light. From these expressions of  $\sigma_{se}$  and  $\beta$  for bulk systems, we find that  $c\sigma_{se}/n_{eff}V = \beta/\tau_{spont}$ . This relationship is used to replace the typical stimulated emission terms involving  $\sigma_{se}$  with  $\beta\Gamma_s\phi_s(N_2 - N_1)/\tau_{spont}$  in our rate equations. The term is directly proportional to the field intensity (through  $\phi_s$ ) as well as the population difference between the two levels connecting the transition (the energy separation between  $N_2$  and  $N_1$  is assumed to be  $hf_l$ ). Such formulation of the stimulated emission term is consistent with those presented by Björk *et al.*<sup>30</sup>. The cavity enhancement effects are then included in our model by replacing  $\beta/\tau_{spont}$  with the modified spontaneous emission rate defined in Eq. (1).

*Intersystem crossing and triplet-state relaxation.* In the rate equations,  $\tau_{isc}$  is the intersystem-crossing lifetime of the molecules from  $S_1$  to  $T_1$ , while  $\tau_t$  is the triplet decay lifetime (both radiative and non-radiative) of molecules from  $T_1$  to  $S_0$ . Phosphorescence arising from the decay of triplet molecules in  $T_1$  is usually overshadowed by fluorescence in  $S_0S_1$  (i.e.  $\tau_{spont} \ll \tau_t$ ) and is not considered in our model. The transitions of molecules between the singlet and triplet states are energetically allowed but spin-forbidden, and so, they occur at a slower rate ( $> 10^{-7}s$ ) than the singlet-singlet transitions ( $< 10^{-8}s$ ). This allows the triplet influences in an organic laser system to be minimized by pumping with a pulse with width shorter than  $\tau_{isc}$ , or by circulating the organic dye solution through the cavity at a rate faster than  $1/\tau_{isc}$ . It is critical to avoid a build up of the triplet population which tend to degrade the lasing performances by increasing the threshold and bleaching rate, while simultaneously decreasing the slope efficiency (see Sec. II of the main text).

*Photobleaching.* Photobleaching of the organic medium is modeled through the irreversible loss of molecules in the triplet state<sup>23</sup>, which is typically regarded as unstable due to its high chemical reactivity in an oxygen environment<sup>24,25</sup>.  $\tau_{bleach}$  is the photobleaching lifetime in  $N_4$ . It can be markedly different depending if the medium is exposed to an oxygen-rich or oxygen-deficient surroundings, or when different solvents are used with the dyes. It is sometimes also modeled to occur in the excited singlet state  $S_1$ <sup>25,31</sup>. In this work, we consider the conventional approach of modeling the loss of molecules in  $T_1$  but note that the model can be extended to also

include the photobleaching process in  $S_1$ .

## APPENDIX B: PARAMETER VALUES

Table I provides the parameters used in the steady-state calculations of Sec. III based on solutions of R6G<sup>42,47,48</sup> (laser wavelength  $\lambda_l = 580 \text{ nm}$ , pump wavelength  $\lambda_p = 532 \text{ nm}$ ):

Dye concentration	$5 \text{ mM}$
Fluorescence quantum yield, $q_{\text{dye}}$	$0.9$
Spontaneous emission lifetime, $\tau_{\text{spont}}$	$5 \text{ ns}$
Intersystem crossing lifetime, $\tau_{\text{isc}}$	$100 \text{ ns}$
Triplet to singlet relaxation lifetime (quencher), $\tau_t$	$100 \text{ ns}$
Triplet photobleaching lifetime, $\tau_{\text{bleach}}$	$8 \text{ ms}$
Non-radiative relaxation lifetime from $i$ to $j$ , $\tau_{ij}$	$1 \text{ ps}$
Self-absorption cross-section in $S_0S_1$ , $\sigma_{\text{abs}}^{S_0S_1}(\lambda_l)$	$10^{-19} \text{ cm}^2$
Self-absorption cross-section in $S_1S_2$ , $\sigma_{\text{abs}}^{S_1S_2}(\lambda_l)$	$10^{-17} \text{ cm}^2$
Self-absorption cross-section in $T_1T_2$ , $\sigma_{\text{abs}}^{T_1T_2}(\lambda_l)$	$10^{-17} \text{ cm}^2$
Thickness of gain region	$30 \text{ nm}$
Confinement factor of the lasing mode, $\Gamma_s$	$0.2$
Spontaneous emission modification factor, $F_p$	$1$
Spontaneous emission coupling factor, $\beta$	$1 \times 10^{-4}$
Total quality factor of passive cavity, $Q_{\text{tot}}$	$5 \times 10^4$
Quality factor of cavity due to radiation-loss, $Q_{\text{cav}}^{\text{rad}}$	$1 \times 10^5$

TABLE I: Parameters of organic laser system based on R6G solution.



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