Optical Phenomena and Dynamics in Organic Microcavity Laser

***Ofer Shapira*[1], *Song-Liang Chau*[1], *Bo Zhen*[1], *and Marin Soljacic*[1]**

[1]Research laboratory of electronics, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA, USA, ofers@mit.edu

Abstract: Formalism for the interaction of organic molecules with a microstructure cavity is presented. We study the effect of emission and absorption properties of organic molecules in a micro-structured cavity on lasing condition. The formalism is based on coupled-mode theory and provides analytical expression for steady state operation. This study suggests routes to obtain an ultra-low threshold organic laser that can ultimately pave the way to development of an electro-luminescent solid-state organic laser device.

While organic light-emitting diodes improved significantly over the past decade and optically pumped organic semiconductors lasers were demonstrated with threshold of only a few tens of W/cm^2 [1], the realization of electro-luminescent (EL) organic laser has remained elusive. An EL organic laser device is attractive due to the high wavelength tunability in the visible afforded by organic molecules, its cost-effective fabrication methods, and improved thermal stability compared to inorganic counterparts [2]. The main hurdles in obtaining EL lasing with organic molecules are the characteristics nanosecond spontaneous lifetime, and the losses associated with excited states absorption, injected charges and contacts, all resulting in relatively high threshold. These properties, together with the low mobility of organic semiconductors that limits the maximum continues current density to a few kA/cm^2,[3] make it extremely challenging to obtain lasing. Although organic materials research has demonstrated improvement in charge mobility [4], the equivalent current calculated from the lowest demonstrated threshold of optically pumped organic semiconductor lasers are still too high. In this work, we have developed a semi-analytical couple-mode theory formalism for the interaction of an organic semiconductor with a cavity, and studied methods to reduce laser threshold to below that required for electrically pumped lasers.

Controlling the spontaneous and stimulated emission of molecules by embedding them into a nano-structured dielectric is an attractive way to tailor the lasing properties of active media. Photonic crystal cavities can localize light in nanoscale volumes with high quality factors, allowing strong interactions between light and matter. As a result, by tailoring the local density of states, fluorescence can be enhanced or inhibited, providing control over the radiative lifetime experienced by a molecule inside the cavity. Here we describe the interaction between the organic molecule and cavity by means of coupled-mode theory. Using first order perturbation theory in Maxwell's equations, one can obtain simple first-order differential equations for the time evolution of the average population densities [5]. Thus, starting from the knowledge of the electric field profile of the resonant cavity and the organic molecule electrical and optical properties, one can compute all the relevant physical quantities characterizing lasing action in such structure by solving the linear system of first-order differential equations. We present formalism that allows us to explicitly retrieve analytic expressions for the lasing threshold and slope efficiency. As a general energy-level model of an organic molecule, we consider the three lowest singlet and two lowest triplet states. Radiative processes are characterized by their lifetimes and emission and absorption cross sections. Transition rates of non-radiative processes, such as internal conversion and intersystem crossing that occurs due to vibrational relaxation, are also considered. We provide a steady state solution to the couple mode equations and study the threshold behavior as a function of the material and cavity parameters.

[1] A. Rose, Z. Zhu, C. F. Madigan, T. M. Swager, and V. Bulovic "Sensitivity gains in chemosensing by lasing action in organic polymers" *Nature*, **434**, April 2005, pp. 876
[2] C. Pflumm, C. Karnutsch, M. Gerken, and U. Lemmer, "Parametric Study of Modal Gain and Threshold Power Density in Electrically Pumped Single-Layer Organic Optical Amplifier and Laser Diode Structures" *IEEE Journal of quantum electronics*, **41**, March 2005, pp. 316
[3] I. D. W. Samuel and G. A. Turnbull, "Organic Semiconductor Lasers" *Chem. Rev.*, **107**, 2007, pp. 1272
[4] B. K. Yap, R. Xia, M. Campoy-Quiles, P. N. Stavrinou, and D. D. C. Bradley, "Simultaneous optimization of charge-carrier mobility and optical gain in semiconducting polymer films" *Nature Materials*, **7**, May 2008, pp. 376
[5] S. L. Chua, Y. Chong, A. D. Stone, M. Soljacic, and J. Bravo-Abad "Low-threshold lasing action in photonic crystal slabs enabled by Fano resonances" *Optics Express*, **19**, 2011 , pp. 1539