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Lasing through a strongly-coupled mode by intra-cavity pumping

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Abstract: We demonstrate room temperature lasing through the polaritonic mode of a J-aggregate microcavity in which losses from exciton-exciton annihilation and slow polariton relaxation typical of direct J-aggregate excitation are circumvented via intra-cavity pumping. The pumping scheme utilizes an organic dye layer (DCM) within the cavity with an emission band overlapping the entire lower J-aggregate polariton branch spectrum, hence forcing DCM lasing to occur through the strongly-coupled mode. This cavity architecture, which separates strong coupling and gain into two materials, presents a general and flexible design for polariton devices and allows for the use of a wide range of materials, organic and inorganic, to be integrated into the cavity.

OCIS codes: (140.3945) Microcavities; (160.4890) Organic materials; (240.5420) Polaritons.

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Strong coupling of light and matter in planar microcavities enables the studies of polariton condensation, superfluidity and related condensed matter phenomena, and opens a path to a radically new class of optoelectronic devices based on the macroscopic coherence of light and matter [1–5]. Notably, strong coupling in molecular organic materials [6,7] could enable lasing and condensation to be achieved at room temperature, as the high absorption constant of many molecular films and crystals can lead to pronounced manifestations of strong coupling even for low quality microcavity structures. Previous work has shown the first polariton laser based on organic materials using crystalline anthracene [8,9]. However, the anthracene laser, as well as other attempts at achieving polariton lasing using J-aggregates as the strong-coupling material, have all encountered the phenomenon of exciton-exciton annihilation as a significant loss mechanism [10]. Exciton-exciton annihilation [11,12] in polariton microcavities is a competing process with the buildup of a threshold population of polaritons at the $k = 0$ point of the dispersion [Fig. 1(a)]. Furthermore, due to the slow exciton-phonon scattering rate and the short polariton lifetime, only a small fraction (10^{-3}) of the photogenerated excitons become cavity polaritons [13,14]. Here we demonstrate a new approach to populating the lower polariton (LP) branch of a microcavity that circumvents losses due to exciton-exciton annihilation [15] and the small polariton fraction, and provides a flexible design architecture for organic and hybrid organic-inorganic polariton devices.

The traditional non-resonant pumping scheme for populating the LP branch involves off-normal-angle excitation of the polariton microcavity with incident light of photon energy well above the LP energy [Fig. 1(a)] [16]. The photogenerated hot excitons then relax to the exciton reservoir and subsequently relax to the bottom of the LP branch by polariton-polariton scattering or phonon-polariton scattering. In contrast, in the intra-cavity pumping scheme [Fig. 1(b)], described in this work, a second, emissive material inside the cavity acts as the LP pump [15]. The broadband emission spectrum of the pump material is chosen to overlap with the entire dispersion of the LP branch. Therefore, any cavity emission from the pump material, either spontaneous or stimulated, occurs through the strongly-coupled LP mode. By utilizing this intra-cavity pump scheme, scattering from the exciton reservoir can be avoided, hence reducing the amount of exciton-exciton annihilation. Furthermore, if the intra-cavity pump material has a large stimulated emission cross-section, lasing of the pump material will occur through the strongly coupled mode, thus not requiring LP-LP scattering to create lasing from a polaritonic cavity. In particular, the four-level structure of organic materials is responsible for low (~10 μJ/cm^2) lasing thresholds [17] [18], which suggests that similarly low thresholds should be possible from a cavity in strong coupling. Lasing through the strongly coupled mode has been suggested to explain the room temperature polariton lasing in anthracene [19]. Regardless of the mechanism of amplification—LP-LP scattering or stimulated emission—the result is coherent emission through a strongly-coupled mode with a single nonlinear threshold. In the case of organic microcavities, the use of two optically active

materials creates a cavity architecture that relaxes the stringent material requirements to achieve organic lasing in a strongly coupled mode by employing one strongly coupled material to create the polariton mode and another material to populate the LP branch and creating lasing.
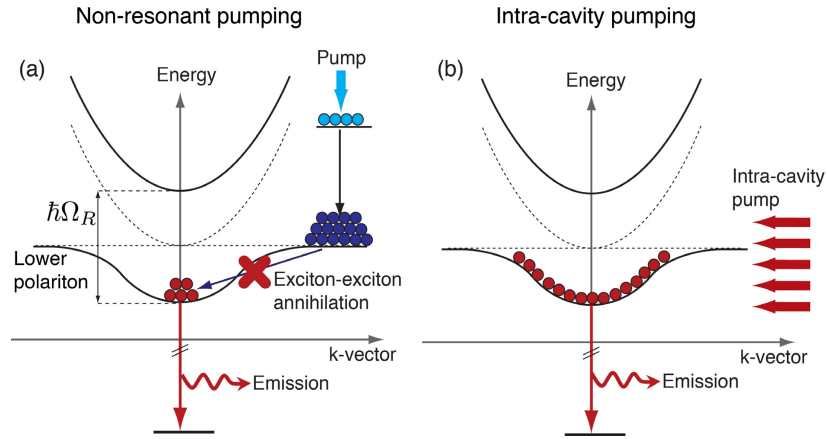

Fig. 1. (a) Non-resonant excitation scheme for polariton microcavities showing exciton-exciton annihilation as a lossy process. (b) Intra-cavity pumping scheme utilizing broadband emission from a second organic material in the cavity to pump the entire LP branch thereby removing the need for polariton-polariton scattering to populate the bottom of the LP dispersion.

In this work, the strongly coupled material is a highly optically absorbing 5-nm thick J-aggregate thin film with an absorption line centered at energy $E = 2.100$ eV (corresponding to the wavelength $\lambda = 591$ nm), with a linewidth of 87 meV, and a peak absorption coefficient of 4×10^5 cm^{-1}. The intra-cavity pump material is the laser dye DCM (4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran), with broadband emission centered at $E = 2.03$ eV ($\lambda = 612$ nm), which overlaps well with the entire LP branch [Fig. 2(b)]. The absorption of DCM is negligible at the LP energy [Fig. 2(b)]. The cavity is fabricated on a quartz substrate by first depositing a 7.5 bilayer SiO_2/TiO_2 distributed Bragg reflector (DBR) by RF magnetron sputtering. A $\lambda_0/4$ SiO_2 spacer layer is deposited on the DBR (where $\lambda_0 = 605$ nm is the average position of the lower polariton branch across the sample) in order to position the subsequently deposited J-aggregate layer at the anti-node of the cavity electric field. The J-aggregate thin film is grown by sequential immersion of the sample into solutions containing the anionic cyanine dye TDBC (5,6-dichloro-2- [3-[5,6-dichloro-1-ethyl-3-(3-sulfopropyl)-2(3H)-benzimidazolidene]-1-propenyl]-1-ethyl-3-(3-sulfopropyl) benzimidazolium hydroxide)) and the cationic polyelectrolyte PDAC (poly(diallyldimethylammonium chloride)) [20]. The remainder of the λ-thick cavity is filled with DCM doped at 2.5% w/w in Alq_3 (aluminum tris(8-hydroxyquinoline)) [21] deposited by thermal co-evaporation. In addition, a 15 nm spacer layer of Alq_3 containing no DCM is deposited on the J-aggregate film to avoid Förster resonant energy transfer (FRET) between the TDBC and the DCM. The top mirror is a thermally evaporated film of Ag with a thickness of 300 nm. The 30 nm effective optical path length at the Ag mirror due to phase shift upon reflection is taken into account in the cavity design. The Alq_3:DCM layer is grown with a spatial gradient to achieve a variable cavity thickness, and hence cavity-exciton detuning, across the sample, with a detuning of 0 meV at the center of the sample. The complete microcavity structure is shown in Fig. 2(a).

The cavity is excited with near transform-limited $\lambda = 400$ nm, 100 fs pulses with a repetition rate of 1 kHz focused through a 0.7 NA microscope objective to a spot size of 20 μm in diameter. The excitation is linearly polarized. The $\lambda = 400$ nm excitation creates Alq_3 excitons which undergo an efficient Förster-resonant-energy-transfer (FRET) to the DCM molecules [21]. Photoluminescence (PL) is collected from the sample through the same

objective with the Fourier plane of the objective imaged onto a fiber coupled to a spectrograph. The fiber is scanned across the momentum space image to obtain the PL dispersion of the cavity with 0.5° angular resolution. Alternatively, an imaging CCD is positioned at the same image plane to obtain the momentum space image [22]. All experiments are performed at room temperature in ambient atmosphere. Angle-resolved PL is shown in Fig. 2(c) for three cavity-exciton detunings (Δ = 10, −50, −90 meV) corresponding to three points on the surface of the sample. A fit of the dispersion to the polariton two-level model results in a Rabi splitting of 60 ± 5 meV and demonstrates that the cavity is in strong coupling.
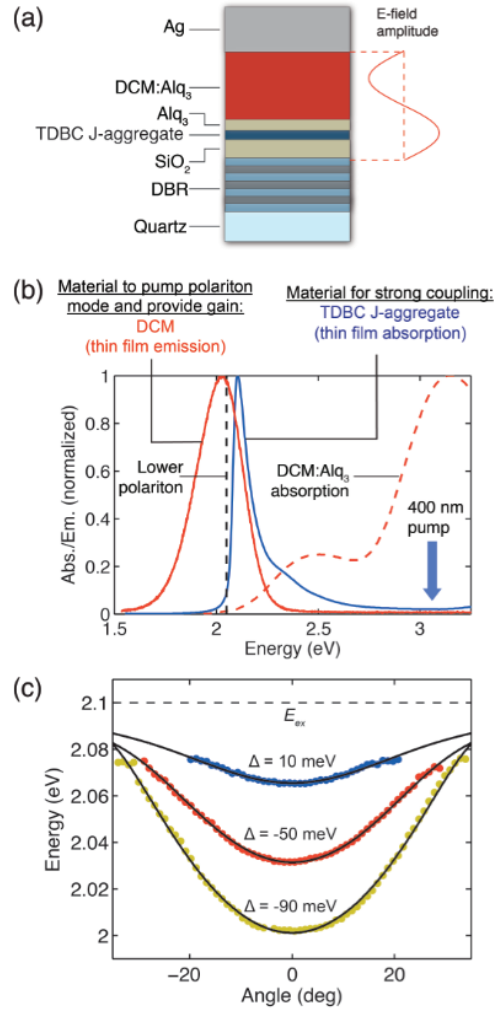

Fig. 2. (a) Schematic of the cavity structure along with an approximate representation of the cavity electric field with the TDBC J-aggregate film at one of the cavity antinodes. (b) Absorption spectrum of TDBC J-aggregates and the emission spectrum of DCM showing overlap with the LP energy. The DCM:Alq₃ absorption is negligible at the LP energy. (c) PL of the LP branch as a function of angle for three cavity-exciton detunings, showing a fit to the LP dispersion, demonstrating that the cavity is in strong coupling.

Figure 3(a) shows the two-dimensional PL dispersion of the polariton cavity with negative cavity-exciton detuning (Δ = −43 meV) under low excitation pulse energy (3.5 μJ/cm^2). At this detuning, the exciton and photon fractions are 0.15 and 0.85, respectively. The LP linewidth is 17 meV corresponding to a polariton lifetime of 40 fs. The LP linewidth is

determined primarily by the J-aggregate homogeneous and inhomogeneous broadening and is not further broadened by the minimal residual absorption of DCM at the cavity resonance energy. The momentum space distribution of the PL shows a wide ± 20° cone emission profile with no observed linear polarization.
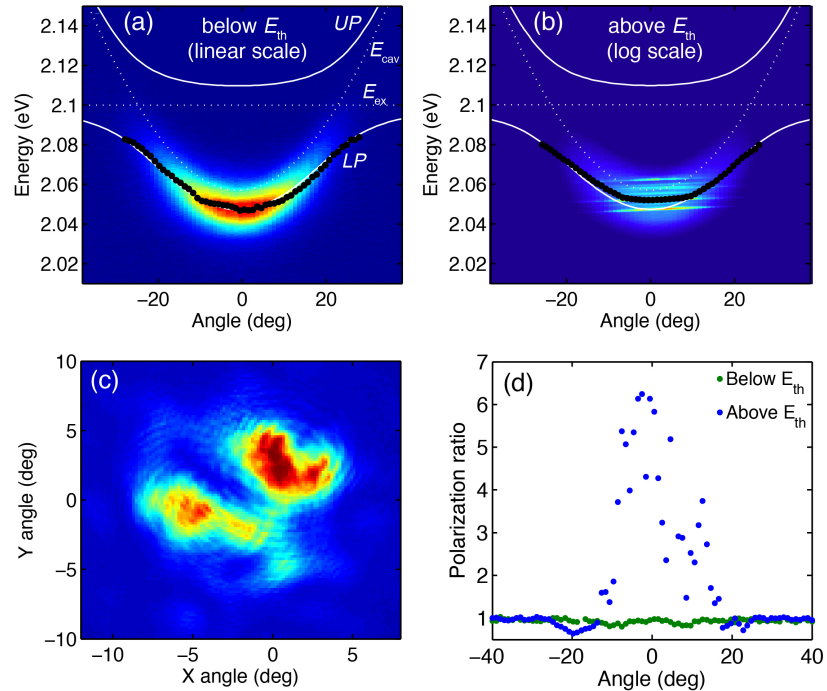

Fig. 3. (a) Photoluminescence dispersion from a cavity with −43 meV detuning pumped below the lasing threshold, E_{th} (shown in linear scale). The plot shows the LP emission maximum at each angle (black circles) and a fit to the LP and UP energy (white solid lines), as well as the energy dispersion of the uncoupled cavity, E_{cav}, and the uncoupled exciton energy, E_{ex} (white dashed lines). (b) Dispersion of same cavity above the lasing threshold, with the intensity shown in logarithmic scale to emphasize that cavity remains in strong coupling above threshold based on the median energies of emission that is not part of the lasing lines (black dots). (c) Emission in momentum space above threshold. (d) Degree of polarization of the emission as a function of angle above and below threshold.

Under increasing pump energy (7 μJ/cm^2), a collapse in the spectral energy width and momentum dispersion of the polariton emission is observed for the cavity with −43 meV detuning (0.15 exciton fraction) [Fig. 3(b)]. Appearance of multiple narrow linewidth modes with a flat dispersion is indicative of the multimode lasing. The non-lasing emission follows the same polariton dispersion as below threshold, with the lasing lines superimposed in energy onto this polariton mode, indicating that the cavity remains in strong coupling above the lasing threshold. The relative intensity of the multiple lasing modes varies with power, but the energy spacing is nearly uniform at ~2.5 meV. Above the lasing threshold, the momentum space distribution shows a narrow ± 5° emission cone angle. The multiple emission spots produce interference fringes in the overlapping regions in momentum space, indicating that the regions in momentum space are coherent with each other [Fig. 3(c)]. A weak coupling cavity with only the DCM gain layer and no J-aggregate layer showed only a single lasing mode. In the strongly coupled cavity, the emission above threshold shows a high degree of linear polarization (ratio of 6) along the direction of the pump laser polarization, despite the FRET that occurs between Alq$_3$ and DCM molecules, behavior which is also observed in DCM microcavity lasers in weak coupling [23]. Due to the amorphous nature of the organic materials, a preferred emission polarization is expected to be set by the pump polarization and

not by the cavity structure. The emission in the center of the lasing cone is the most polarized, with spontaneous non-lasing emission at higher angles showing no linear polarization [Fig. 3(f)].
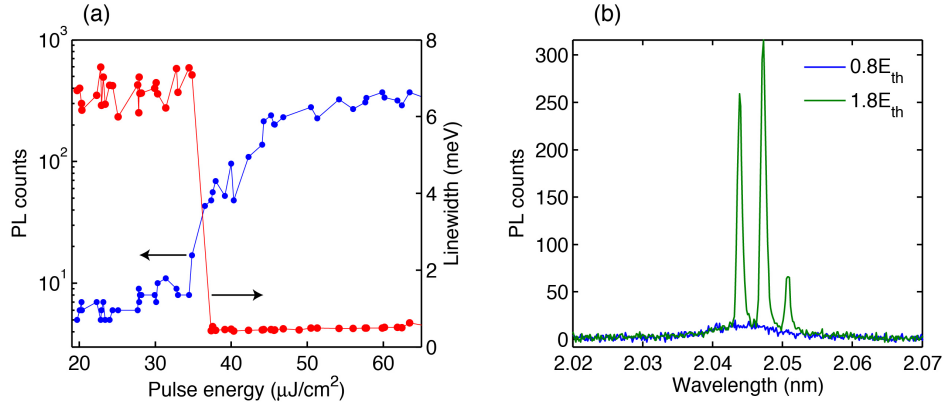

Fig. 4. (a) Dependence of PL emission intensity at $k = 0$ as a function of the excitation pulse energy showing a lasing threshold at 6 μJ/cm². A reduction in the emission linewidth from 7 meV to 0.5 meV is observed at the lasing threshold. (b) Emission spectrum below ($0.8E_{th}$ excitation energy) and above ($1.8E_{th}$ excitation energy) the lasing threshold.

The threshold of nonlinear emission occurs at an absorbed excitation energy density of 6 μJ/cm², accompanied by a narrowing of the emission line from 17 meV (below threshold) to 0.5 meV (above threshold) [Fig. 4]. Saturation of the emission occurs at 8 μJ/cm². We note that the threshold occurs at an energy density below the onset of exciton-exciton annihilation [10], which we found to be ~10 μJ/cm². This input-output dependence is similar to what is observed in DCM VCSELs in weak coupling but the threshold for the polariton cavity at −43 meV detuning is ~2-fold higher than the threshold we find for a DCM VCSEL due to losses in the polariton mode. A nonlinear lasing threshold occurs for polariton cavities with detunings ranging from −35 meV (0.15 exciton fraction at $k = 0$) to −110 meV (0.04 exciton fraction at $k = 0$), with a lower lasing energy threshold for more negative detunings. No threshold was observed for detunings of less than −35 meV due to J-aggregate photobleaching on the time scale of measurement time (~30 s). In addition, for detunings of less than −35 meV, an increased exciton fraction results in higher nonradiative loses due to the increasing J-aggregate exciton portion of the exciton-polariton, which was measured to have a PL quantum yield at room temperature of $(10 \pm 2)\%$. The multiple spectral lasing modes were only observed for cavities containing J-aggregates, at all detunings, and not for weak coupling cavities containing DCM. The relative intensity of the multiple spectral peaks varies with increasing excitation energy, with a single mode dominating at higher power. The multiple modes can likely be attributed to disorder in the J-aggregate film across the excitation spot, an effect that is also observed in GaN polariton lasing [5].

In conclusion, we demonstrate lasing through a strongly coupled mode, achieved by intracavity pumping of a J-aggregate organic microcavity at room temperature. The laser shows spectral and momentum space collapse of the emission above threshold while the cavity remains in strong coupling. The laser employs a new architecture in which the strong coupling material is separated from the material that populates the lower polariton branch and creates gain in the cavity. This architecture opens the possibility for building lasers operating through a strongly-coupled mode at a wide range of wavelengths (from UV to NIR) simply by choosing a J-aggregating molecule with the appropriate absorption line and a corresponding, spectrally overlapping organic pump material. Furthermore, organic materials such as DCM could be incorporated with inorganic quantum wells to create hybrid polariton structures with intra-cavity pumping [24].

Acknowledgments