Intracavity optical pumping of J-aggregate microcavity exciton polaritons

The MIT Faculty has made this article openly available. Please share how this access benefits you. Your story matters.

<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>As Published</td>
<td><a href="http://dx.doi.org/10.1103/PhysRevB.82.033305">http://dx.doi.org/10.1103/PhysRevB.82.033305</a></td>
</tr>
<tr>
<td>Publisher</td>
<td>American Physical Society</td>
</tr>
<tr>
<td>Version</td>
<td>Final published version</td>
</tr>
<tr>
<td>Accessed</td>
<td>Fri Dec 28 15:19:41 EST 2018</td>
</tr>
<tr>
<td>Citable Link</td>
<td><a href="http://hdl.handle.net/1721.1/60352">http://hdl.handle.net/1721.1/60352</a></td>
</tr>
<tr>
<td>Terms of Use</td>
<td>Article is made available in accordance with the publisher’s policy and may be subject to US copyright law. Please refer to the publisher’s site for terms of use.</td>
</tr>
<tr>
<td>Detailed Terms</td>
<td></td>
</tr>
</tbody>
</table>
We demonstrate intracavity optical pumping of J-aggregate microcavity exciton polaritons. The use of ultrathin layer-by-layer J-aggregate thin films as the strongly coupled exciton medium allows for inclusion of a thermally evaporated luminescent cavity spacer layer, through which the lower-branch exciton-polariton states are resonantly pumped. We present a measurement of the lower-branch exciton-polariton occupation in room-temperature microcavity devices containing J-aggregated molecular thin films under low-density steady-state excitation. The observed exciton-polariton occupation shows a Maxwell-Boltzmann distribution at $T = 300$ K, indicating thermalization of exciton polaritons in the lower energy branch. This device design enables us to propose a new type of “polariton laser” architecture for microcavity exciton polaritons.

**DOI**: 10.1103/PhysRevB.82.033305

**PACS number(s)**: 78.55.Kz, 71.36.+c, 81.05.Fb
The use of a luminescent Alq3:DCM top cavity spacer allows for deposition of J-aggregate thin films using less-water-soluble dyes than THIATS, such as TDBC, the J-aggregating dye used in the electroluminescence studies referenced above.5,29

After the J-aggregate thin film is deposited on top of the bottom DBR, a luminescent top cavity spacer of varying thickness is thermally evaporated, consisting of Alq3 [aluminum tris(8-hydroxyquinoline)] doped at 2% to 3% by weight with the laser dye DCM [4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran]. The varying thickness of the spacer layer across the substrate [as sketched in Fig. 1(a)] is achieved by evaporating the Alq3:DCM thin film through a fixed shadow mask onto a rotating substrate. The hybrid process of SSA and a few LBL immersion steps can also be performed to allow deposition of J-aggregate thin films using less-water-soluble dyes than THIATS, such as TDBC, the J-aggregating dye used in the electroluminescence studies referenced above.5,29

FIG. 1. (a) Graded-thickness microcavity device structure and molecular diagrams of constituent thin film materials. (b) Normalized absorbance (Abs) and photoluminescence (PL) spectra of Alq3, DCM, and THIATS J-aggregates showing spectral overlaps governing energy down conversion from Alq3 through DCM to THIATS J-aggregates.

FIG. 2. (Color online) (a) PL spectrum versus position on sample with energies of lower-branch (LB) and upper-branch (UB) exciton-polariton modes, uncoupled cavity mode found with numerical fit (Cav), and uncoupled J-aggregate exciton (Ex). (b) PL spectrum versus angle at position Y=10 mm on sample using low-resolution grating. (c) PL spectrum versus angle at same position as in (b) using high-resolution grating.

For PL measurements, the samples are pumped using a 4 mW λ=408 nm CW laser focused into a<100 μm diameter spot, which excites the Alq3 and subsequently DCM through Förster transfer; the DCM then excites the THIATS exciton and exciton-polariton states through direct Förster transfer or through absorption of DCM PL by THIATS, depending on the proximity of the excited DCM molecules to the THIATS J-aggregate thin film. Angle-resolved PL is collected using a multimode optical fiber mounted on a custom-built automated rotation stage with <0.5° resolution. The excitation laser strikes the sample at a 45° incident angle; a slight downward tilt to the optic axis is used to avoid collection of the reflected excitation beam by the fiber.

Figure 2 shows the PL intensity collected from the sample shown in Fig. 1(a) as a function of position on the sample surface, with the PL collected at normal incidence (i.e., k|| =0) [Fig. 2(a)] and as a function of angle of light collection away from the normal [Fig. 2(b) and 2(c)]. The exciton reso-
nance (i.e., the ground state to one-exciton transition of the J-aggregate) is assumed to be at a free-space wavelength of $\lambda_{J-Agg} = 630$ nm. The wavelength of the cavity resonance, $\lambda_{ca}$, is calculated at each position by fitting the observed peak in the lower-branch exciton-polariton PL to that predicted by the coupled light and two-level system eigenenergy equation:

$$E_z = \frac{1}{2}(E_{ca} + E_{J-Agg}) \pm \frac{1}{2}\sqrt{h^2 \Omega_R^2 + (E_{ca} - E_{J-Agg})^2}, \quad (1)$$

where $E_{ca}$ are the upper and lower-branch exciton-polariton eigenenergies, $E_{ca}$ is the energy of the cavity resonance corresponding to $\lambda_{ca} = h\omega/\lambda_{ca}$, $E_{J-Agg}$ is the exciton resonance corresponding to the $\lambda_{J-Agg}$, and $\Omega_R$ is the Rabi splitting, with $\Omega_R = 80$ meV used for the fit.  

Figure 2(b) shows the angle-resolved PL at nearly-zero detuning [position $Y \sim 10$ nm on the Fig. 2(a) plot]. Note that the PL intensity in this figure is plotted on a logarithmic scale. PL counts are scaled to correct for the above effects indicated by scaled PL counts versus energy. PL counts are scaled to correct for the above effects. The good agreement between the measured lower-branch exciton-polariton relative occupation and the M-B distribution at $T=300$ K indicates that the exciton polaritons pumped into the lower-branch have thermalized prior to decay.

The measured intensity of the lower-branch exciton-polariton PL, shown in Fig. 2(c), is converted to a relative occupation number of the corresponding exciton-polariton states by taking account of both the effect of the multilayer planar structure on the observed emission intensity and the effect of the changing composition of the exciton polariton from photon to exciton due to the larger positive detuning of the cavity resonance from the exciton resonance as the emission angle increases. The former is an effect of Snell’s law and is corrected by dividing the observed counts by the cosine of the observation angle (as in Lambert’s Cosine Law). The latter is corrected by calculating the Hopfield coefficients at each angle and then dividing the observed counts at each angle by the respective photon fraction. Figure 3(a) shows the observed counts of the lower-branch exciton-polariton PL (scaled to correct for the above effects) together with a superimposed plot of the Maxwell-Boltzmann (M-B) distribution at $T=300$ K. Figure 3(b) shows the photon and exciton fraction of the lower-branch calculated from the Hopfield coefficients and used in the scaling of the counts in Fig. 3(a). The good agreement between the measured lower-branch exciton-polariton relative occupation and the M-B distribution at $T=300$ K indicates that the exciton polaritons pumped into the lower-branch have thermalized prior to decay.

Use of the optically pumped luminescent spacer layer to inject exciton polaritons into the strongly coupled microcavity, which is then followed by the exciton-polariton thermalization in the lower branch, suggests a device architecture for a “polariton laser.” With sufficiently high reflectance of the microcavity mirrors, and a sufficient thickness of the luminescent cavity spacer gain material, the spacer material could undergo lasing in an exciton-polariton mode.

Although exciton-polariton lasers are generally expected to have a lower threshold than traditional vertical-cavity surface-emitting lasers (VCSELs), based on current literature, a comparison of the expected threshold of an organic VCSEL to that of an organic exciton-polariton laser is complicated by the lack of studies specific to organic exciton-polariton systems. Litinskaya et al. estimated a low threshold for organic exciton-polariton lasing relative to organic VCSELs by using the density of states of a planar microcavity to determine occupation threshold, but not accounting for the effect of dephasing on condensation, which has been studied at length in inorganic systems. From the literature on inorganic exciton-polariton systems, one might com-
pare GaN-quantum-well systems to organic systems since both are operated at room temperature and have similar Rabi splittings (30–150 meV) and exciton linewidths (15–50 meV) \(^{33,34}\) but some important distinctions remain. For example, the threshold of a quantum-well GaN-based exciton-polariton system, recently predicted theoretically, was \(3 \times 10^{12}\) excitations per cm\(^2\), which corresponds to one exciton polariton per \(5.7 \times 5.7\) nm\(^2\) \(^{35}\). Such a length scale is comparable to the Förster radius for molecule-to-molecule exciton energy transfer and to the exciton diffusion length of many molecular organic thin films, especially those with considerable spectral overlap of emission and absorption which are typically used in exciton-polariton devices. This proximity in length scale necessitates that dipole-dipole interactions, such as present in exciton energy transfer and exciton-exciton annihilation, must be explicitly taken into account.\(^{36,37}\) For comparison, assuming that a pump photon energy is 3 eV, then \(3 \times 10^{12}\) excitations per cm\(^2\) would correspond to the lasing threshold energy density of \(E_{th} \sim 1\) \(\mu\)J cm\(^{-2}\), which is two orders of magnitude lower than demonstrated for organic VCSELs, to date.\(^{38}\) Note that a high peak pump power \((\sim 1\) MW cm\(^{-2}\)) would be required for exciting the exciton-polariton laser, with even higher power needed than for organic VCSELs, owing to the short photon lifetime of \(\tau \sim 1\) ps in the cavity.\(^{39}\) The short cavity photon lifetime also governs the excited state lifetime of exciton polaritons, which inhibits their diffusion, and inadvertent polariton-polariton annihilation.

In conclusion, we demonstrated intracavity optical pumping of exciton polaritons in J-aggregate microcavity systems using a luminescent cavity spacer layer and show that the occupation of the lower-branch exciton polaritons in such a system at room temperature is thermalized, with relative occupation following a Maxwell-Boltzmann distribution at \(T = 300\) K. We suggest that a type of “polariton laser” could be fabricated in organic systems by making a VCSEL whose gain material is the luminescent cavity spacer layer and which lases in a strongly coupled exciton-polariton mode.

M.S.B. acknowledges the support of the Department of Defense. The work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences as part of the MIT Energy Frontier Research Center on Excitronics.

---

*bulovic@mit.edu