Skoltech
Skolkovo Institute of Science and Technology

Thesis Changes Log

Name of Candidate: Timur Yagafarov

PhD Program: Physics

Title of Thesis: Polariton lasing in a dye-filled organic microcavities

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Chair of PhD defense Jury: Prof. Nikolay Gippius ***Email**: N.Gippius@skoltech.ru*

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The thesis document includes the following changes in answer to the external review process.

Dear Jury Members,

Thanks for your positive reviews and valuable questions on the defense. Please, find here the answers to the question left. I would like to thank you again for a help in my thesis improvement.

Answers to Simone De Liberato:

The discussion about strong coupling criterion have been modified as follows:

The strong coupling criterion mentioned above is rather intuitive than strictly defined. The further clarification of strong coupling condition can be considered from the spectroscopic criterion which says that the measurable splitting of two polariton states can be resolved if the frequency splitting is bigger than the sum of the linewidths of loss rates $\Omega_0 > \frac{|\gamma_1+\gamma_2|}{2}$, according to [34,35]. Sure, that definition is reasonable in terms of the experimental approach. If one wants to access the general criterion for strong coupling, the proper figure of merit would be the ratio $\frac{\Omega_0}{\gamma}$, where γ is the loss rate, and Ω_0 is the vacuum Rabi frequency. If the ratio $\frac{\Omega_0}{\gamma} > 1$, the system appears in the

strong coupling regime, where an efficient energy exchange between oscillators can happen before the losses in the system will lead to the damping of the oscillators.

Answers to Luis Vina:

•In page 29, "…the binding energies comparison above makes it clear why the organic materials are stable …" should read "…the binding energies comparison above makes it clear why the excitons in organic materials are stable …"

The correction was implemented as suggested by reviewer. (Page 29)

• A quote to the work of Hopfield, "Theory of the Contribution of Excitons to the Complex Dielectric Constant of Crystals", Phys. Rev. 112, 1555 (1958), should be given together with Ref. [33].

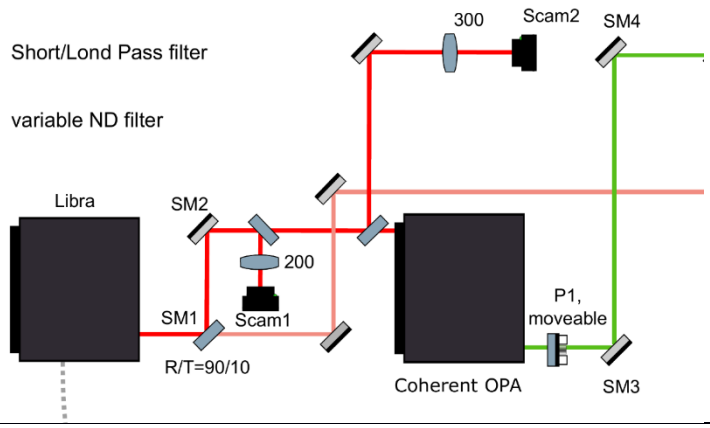
The quote of Hopfield work was added. Now it reads: "…which possess the properties of both the photon and the exciton [33,34]." (Page 30)

• In page 45, the sentence "Thereby, the wavelength of polariton lasing, which expected to emit from the lower polariton state, has an opportunity to be tuned…" is quite strange, it should be re-written.

The sentence has been corrected: "…select preferred energy of the ground polariton state. Thereby, the wavelength of polariton lasing has an opportunity to be tuned." (Page 45)

• In Fig. 3.6 there is a black box that is not identified (the one on the right)

Thanks for the correction! The "Coherent optical parametric amplifier system" has been identified in Figure 3.6 (Page 49).



Additional reference for the Figure 3.6 was made in the description of Z-scan measurement, where we used the Coherent OPA system: "...pumped the optical parametric amplifier (Coherent OPerA SOLO, see in Figure 3.6) and provided 550 nm central emission wavelength…" (Page 69)

• *In page 54, it is written: "In real systems, the threshold for polariton lasing is expected to be less than the estimated value because of scatterings and the leaky tunneling modes [65]…", is ok less or should be more?*

Yes, it should be **less** than estimated value. We estimated the absorbed part of the pump just from the transmission and reflection subtraction from the incident pump. However, in the microcavity we have a leaky tunnelling modes, and scatterings are also possible for the excitation pump. In our rough estimation they are automatically taken into account in the absorption, but if we will consider them, then absorption value will be lower than estimated. Therefore, the threshold will be also lower.

• *In page 58, it is written: "…we were able to measure the energy of polariton lasing, leaving the microcavity, which found at ~5 nJ…", this is grammatically a quite strange construction, it should be re-written. Furthermore, is it 5 nJ or 5 pJ?.*

Thanks for finding a typo. The sentence has been corrected and rewritten in friendly way: "…we were able to measure the energy of polariton lasing, which found at ~5 pJ." (Page 58)

• *Fig. 4.3. Do the data plotted in (b) correspond to those of (a), it does not look like that this is the case. I have added the cyan lines to the graphs, and, for example, at 24º I believe that what is plotted in (b) does not correspond to what is shown in (a).*

Yes, that is true, thanks for the correction. The reflectivity spectra were not the same as on the color plot (they were taken from different points). I changed the reflectivity spectra in Figure 4.3 to appropriate one. Also correction was implemented in the text and the legend. (Page 64).

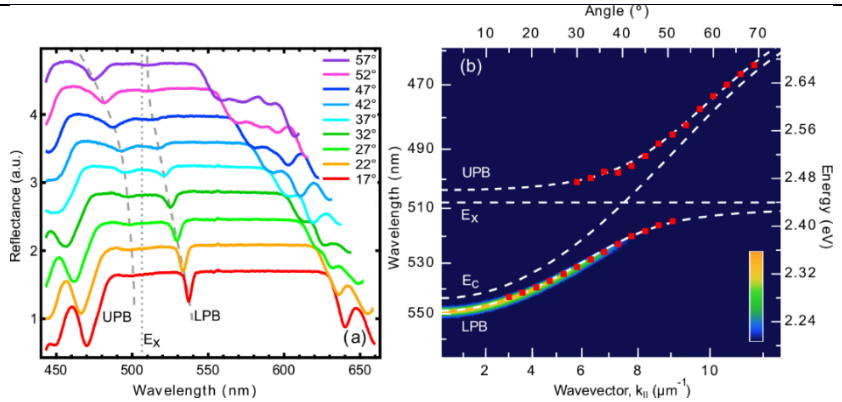


Figure 4.3. Strong light-matter interaction in dye-filled microcavities. (a) Angle-dependent reflectivity spectra of the microcavity recorded at different angles exhibit clear anti-crossing at the exciton resonance energy Ex (grey dotted line) and indicate the formation of lower (LPB, grey dashed line) and upper (UPB, grey dashed line) exciton-polaritons branches. Polariton dispersion relation in (b) is plotted by combing the data of photoluminescence imaging acquired in a Fourier space (rainbow colour density plot in a log scale) with the polariton states extracted from angle-dependent reflectivity measurements (red squares). Fits for the LPB and UPB, together with the cavity mode E_c and energy of exciton resonance Ex are shown as a white dashed curves.

• *After the Eq. 4.2 is written: "...where, Np - polariton and NX - exciton reservoir densities…". What is meant by polariton reservoir?*

I meant polariton density and exciton reservoir density, separately. To avoid further confusions, I have rephrased the sentence: "…N_p – polariton density and N_X - exciton reservoir density." (Page 72)

• *Pg. 104: "To demonstrate the role of the ET in the polariton dynamics…" ET (energy transfer) is not defined. ET appears also as a label in Fig. 4.13.*

Thanks for correction. The definition for "ET" have been introduced in the first mentioning of energy transfer: "…from an interplay between stimulated relaxation to the polariton ground-state and intermolecular energy transfer (ET)." (Page 61)

General comment: In all figures where is used a false-colour-scale the numerical limits of the scale should be given.

Thanks for the comment. Corrections were implemented where appropriate.