Skoltech
Skolkovo Institute of Science and Technology

Thesis Changes Log

Name of Candidate: Anton Putintsev

PhD Program: Physics

Title of Thesis: Ambient Polaritonics

Supervisor: Prof. Pavlos G. Lagoudakis

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

Date of Thesis Defense: 11 November 2022

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

Dear Jury Members,

First of all, I am very grateful for your very constructive feedback and criticism on the overall impression from reading this work and for pointing out particular defects. Narrative seamlessness, consistency of references, figures, chapters, and sections are essential parts of a dissertation. All drawbacks, comments, and suggestions related to the mentioned above have been maintained and amended correspondingly in the final version of the thesis.

Please, find below both a detailed description of modifications made and answers to your questions and comments in the final thesis version. The pages referenced here correspond to the pages from the main work accordingly.

Reviewer: Prof. Daniele Sanvitto

- *Inset of figure 3.2 is useless since it is already in Figure 3.1*
- *Figures 5.1 and 5.2 are absent in the text and figure 5.2 has a different caption*
- *Page 41 there is a cm^1 which is clearly wrong.*

Answer: Corrected.

- *Section 4.1, except maybe the last paragraph, could go to Chapter 2. It doesn't make sense after Chapter 3.*

Answer: First sections of Chapter 3 and Chapter 4 have been revised and partially merged together to avoid contextual overlapping and repetition of some facts, references and information.

- *Page 41 there is a cm^1 which is clearly wrong.*

Answer: Of course, the reciprocal momentum space has a dimensionality of ***[μm^{-1}]*** *and to extract the blueshift and FWHM of the condensate emission, single-shot k-space dispersion images profiles were filtered over* ***±0.075 μm^{-1}*** *range around k = 0.*

- *Page 44 "step size of 1 m" ?*

Answer: Indeed, there is a typo. The photoluminescence (PL) emission was mapped over the area of (26x26) μm^2 with a step size of ***1 µm*** *using a laser beam focused down to 4 µm and motorized XY stage.*

- *Section 5.1, at least the first half, should be rewritten considering it is the 5th chapter of a thesis not the introduction of an article.*

Answer: Thank you for binging my attention on this problem. Section 5.1 of Chapter 5 has been revised and rewritten according to the format of the dissertation. Some of the parts were moved to the introduction section to maintain consistency and continuity of the story.

Reviewer: Prof. Sakellaris Mailis

- *"I found that especially chapter 3 suffers from an infestation of typos which need to be corrected for clarity and the avoidance of confusion. Additionally, I found that the abstract is too vague and does not capture successfully the particulars of this work."*

Answer: I fully agree with raised points. That is why Chapter 3 has been carefully revised. The Abstract part has been also extended to additionally summarize the findings of each chapter. The rest text has been modified accordingly.

Reviewer: Prof. Dmitry Gorin

- *"Main object of research was BODIPY-Br dye molecules. There are many others variants for example J-aggregates of ICG. I did not find enough explanation about this chose in thesis."*

Answer: Thank you for stressing this out. Indeed the choice of BODIPY-Br dye molecules was not discussed in the thesis. In fact, in the previous work of my colleges [1] it was shown that boron-dipyrromethene fluorescent dye (BODIPY-G1) can generate polariton lasing at room temperature over a broad spectral range. Nevertheless, demonstration of polariton lasing in the yellow part of the spectrum in an organic microcavity containing the molecular dye bromine-substituted boron-dipyrromethene (BODIPY-Br) [2] proved this material to be more photostable and, hence, more successful for studying polariton physics under a longer excitation pulses and at higher fluencies. BODIPY-Br just considered to be a better candidate and sort of a second generation material in the long pursuit for the best organic polariton platform.

- *"The author wrote: we restrict the range of pump influences to avoid any possible degradation of the structure. Would be useful to consider a possible photodegradation of BODIPY molecules during irradiation using pulse laser;"*

Answer: Thank you for bringing this issue that the reader might find confusing while reading this part. In fact all the measurements in this part of the work were carried out in in a single-shot regime. Since the BODYPY molecules as an active medium for polariton applications is a very sensitive and gentle material, it cannot operate or undergo polariton condensation under CW excitation. In fact, to the best of my knowledge, none of the organic platforms to date have been shown to exhibit polariton condensation in a CW regime. For this particular reason, the single-shot excitation regime is eventually is the only option to go by. While it helps to neutralize the effect of pump power fluctuations, it also allows one to avoid a rapid photodegradation of the material, as usually depending on the pulse duration BODIPY can sustain only

finite number of pump pulses. Additionally, reducing the peak fluency of each pulse helps further to mitigate the photodegradation effect.

- *"I did not find any explanation about the thickness of silver coating. It was equal to 200 nm."*

Answer: The choice of silver mirror thickness of 200nm was not completely arbitrary. Simulations of 1-pair and 5-pair hybrid cavities, ***Fig.4-5*** *from the amended Thesis on p.38, show the penetration of the field into the different mirrors and the refractive index variation.* ***Fig 4-4b*** *shows with a black curve the modelled penetration depth of the electromagnetic field into the bottom mirror in each structure. For the H1 cavity that was primarily the subject of investigation the penetration depth is less the 70nm, hence 200nm silver mirror excludes any boundary effects and is simply easier to be spin-coated.*

- *"Page 38, Figure 4-4, Why is the experimental quality factor higher compared to the model quality factor after 3 pairs of hybrid mirrors?"*

Answer: Fig.4-4c shows how the linewidth of the normal incidence LPB emission depend on the number of DBR pairs on top of the silver mirror in both the experiment and theory. We can see that the experimental linewidth never exceeds theoretical linewidth, Hence, in model the quality factor is always higher compared to one in the experiment for the same structures. Fig.4-4d with a blue solid line shows how Q-factor of a hybrid metal-DBR cavity depends on the number of DBR pairs on top of the silver mirror, while blue dots represent a constant Q-factor of a single control DBR-DBR cavity. It does not show any dependence, but modelled Q-factor of the DBR control cavity.

- *"A summary after every chapter including the review chapter would be very useful."*

Answer: A summary after each Chapter was incorporated into the corresponding discussion Section that summarizes main findings and gives some prospects and personal outlook of the Author.

Reviewer: Prof. Simone De Liberato

- *"On page 28, "Onc" should be "One"."*
- *"In Figure 5-9 there is a "-1" missing in the axis of the bottom-left panel."*

Answer: Corrected.

- *"The last Chapter could stress a bit more the personal outlook of the Candidate."*

Answer: The conclusions section has been expanded in order to elaborate the author's opinion.

References

[1] 1. D. Sannikov, T. Yagafarov, K. Georgiou, A. Zasedatelev, A. Baranikov, L. Gai, Z. Shen, D. Lidzey, and P. Lagoudakis, Room Temperature Broadband Polariton Lasing from a Dye-Filled Microcavity. Advanced Optical Materials, 7, 1900163 (2019)
[2] T. Cookson, K. Georgiou, A. Zasedatelev, R. T. Grant, T. Virgili, M. Cavazzini, F. Galeotti, C. Clark, N. G. Berloff, D. G. Lidzey, and P. G. Lagoudakis, "Polariton condensates: A yellow polariton condensate in a dye filled microcavity" Adv. Opt. Mater. 5, 1700203 (2017).