

Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Anton Putintsev

PhD Program: Physics

Title of Thesis: Ambient Polaritonics

Supervisor: Professor Pavlos Lagoudakis

Co-supervisor: Dr. Denis Sannikov

Name of the Reviewer: Daniele Sanvitto

I confirm the absence of any conflict of interest (Alternatively, Reviewer can formulate a possible conflict)	Date: 04-10-2022
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The purpose of this report is to obtain an independent review from the members of PhD defense Jury before the thesis defense. The members of PhD defense Jury are asked to submit signed copy of the report at least 30 days prior the thesis defense. The Reviewers are asked to bring a copy of the completed report to the thesis defense and to discuss the contents of each report with each other before the thesis defense.

If the reviewers have any queries about the thesis which they wish to raise in advance, please contact the Chair of the Jury.

Reviewer's Report

Reviewers report should contain the following items:

- Brief evaluation of the thesis quality and overall structure of the dissertation.
- The relevance of the topic of dissertation work to its actual content
- The relevance of the methods used in the dissertation
- The scientific significance of the results obtained and their compliance with the international level and current state of the art
- The relevance of the obtained results to applications (if applicable)
- The quality of publications

The summary of issues to be addressed before/during the thesis defense

The thesis “Ambient polaritonics” by Anton Putintsev concerns with the study of condensation of organic microcavity polaritons at room temperature. In a first part of the thesis the condensate dynamics of an organic microcavity is studied via single shot measurements and long temporal pulse excitation. Condensation is also reached via a novel hybrid metal-DBR mirror device that allows for a wider stopband and better or similar Q factors than those of standard microcavities fully made of DBRs. The second part of the thesis is dedicated to the study of the second-order coherence function in an organic microcavity working at room temperature. However rather than the standard HBT setup for the observation of correlations here a different approach is used. Indeed correlations are extracted via a stochastic analysis having access to the intensity of the condensate and its fluctuations. This was possible thanks to a single-shot detection technique reasonably put in place during the PhD studies. In details, this thesis is composed by three main chapters after a first one used as an overview of the work. Chapter 2 is an introductory chapter of polaritons and their ability to condense in microcavities describing the organic materials used for room temperature condensation. Chapter 3 describes the formation and decay dynamics of a polariton condensate in a $\lambda/2$ microcavity filled with BODIPY upon a 4ns pulsed excitation. The measurements show all the characteristics of a condensation including a narrowing of the emission area. It is interesting to see how the condensed state follow the long excitation pulse yet with a shorter lifetime. In Chapter 4 a comparison between a hybrid metal-DBR-cavity-DBR, with a reduced pairs in the bottom DBR, and a DBR-cavity-DBR with 10 pairs each is made. A lower threshold is found for the latter structure while the former may give better Q factors and wider stopbands at the expenses of a worse spatial homogeneity. Chapter 5, which is the last chapter of this thesis, describes a photon statistical analysis of a condensate of polaritons obtained by using a $\lambda/2$ microcavity with a film of MeLPPP sandwiched between two DBR mirrors. One of the key results is the observation that, despite the room temperature operation and the unavoidable inhomogeneities of organic materials, a strong reduction of fluctuations is observed in the second-order correlation function. In particular this tendency is more pronounced, and condensation threshold lowered, for higher area condensates for which the escape of polaritons from the excitation spot is minimized. These observations are backed up by a theoretical model and numerical analysis of the system under study.

I appreciate the work done, as well as the setup put in place for the statistical analysis. In particular it is quite interesting the detail analysis of the condensate dynamics of organic polaritons as well as the study of the hybrid metal-DBR microcavity which could lead to a reduction on the fabrication time and resources. The study of the second-order correlation function is also very well done using a statistical approach which can lead to straightforward results without the need of cumbersome photon correlation techniques that could require long acquisition time and dedicated setups. Surprising is also the observation of a reduction in the fluctuation of the organic-based condensate despite the very disordered composition of the active layer as well as the amorphous nature of the rest of the structure.

These results are in line with the advances of the polariton community as demonstrated by the articles published on this subject and there is a significant degree of interest in relation to future realization of room temperature polariton devices.

The candidate has also published a few good papers and a Physical Review X is currently under review. To note that he is almost always either the first or second author of these publications suggesting that he was the main responsible of the work done, including the experimental setups.

Having said that I cannot but notice that most of this thesis is a cut and paste from the articles published or in preparation. In general, although I do not disagree in using the structure and some wording of the works done during the thesis period, the unity of the dissertation should be maintained and care should

be made that all the references, including figures, equations, chapters and sections are pointing to right item. Here not only many of the references have the numbering of the original paper but sometimes there are missing parts, given there are still references to Supplementary materials and figures which are inexistent. I find this very serious because it means that the student has written his thesis with little care. Careless to the point of not even reading it once, as it is very easy to spot these trivial mistakes.

It is therefore imperative that a strong revision is made to the whole dissertation before the final viva.

Amongst the corrections it could be useful to take care also to these problems:

- Inset of figure 3.2 is useless since it is already in figure 3.1
- Section 4.1, except maybe the last paragraph, could go to chapter 2. It doesn't make sense after chapter 3
- A zoom, as an inset, of the high reflectivity of the different devices in figure 4.1 (c) could help
- Page 41 there is a cm^1 which is clearly wrong.
- Page 44 "step size of 1 m" ?
- 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.
- Figures 5.1 and 5.2 are absent in the text and figure 5.2 has a different caption
- Formula 5.8 is not existent.

Provisional Recommendation

I recommend that the candidate should defend the thesis by means of a formal thesis defense

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The thesis is not acceptable and I recommend that the candidate be exempt from the formal thesis defense