

Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Timur Yagafarov


PhD Program: Physics

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

Supervisor: Prof. Pavlos Lagoudakis

Date of Thesis Defense: 8 November 2019

Name of the Reviewer: Prof. Simone De Liberato

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| I confirm the absence of any conflict of interest | Signature:  Date: 21-09-2019 |
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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

In this thesis the candidate describes the realization and experimental characterization of a broadband organic polariton condensate. Such a condensate is then exploited to investigate the origin of the condensate blueshift universally observed in organic microcavities, whose origin was until now unclear.

The structure of the dissertation is simple and clear. It starts with a brief introduction describing light-matter coupling in organic microcavities, followed by a chapter describing the achievement and characterization of the broadband organic condensate. The fourth chapter contains the attempt, both theoretical and experimental, to identify the mechanism behind the condensate blueshift. A very short outlook chapter closes the manuscript.

Although the linguistic skills of the candidate are not the main object of evaluation here, I have to point out that the level of the English presentation is insufficient, as at times it makes the manuscript very difficult to understand, thus strongly limiting its scientific value. The whole dissertation has to be checked and the language significantly improved before the viva takes place.

The scientific methodology used through the paper is sound, and the result obtained are both of the highest quality and very timely. I particularly appreciated the thorough investigation of the different possible mechanisms behind the condensate blueshift. Overall this thesis strongly advances the state of the art in the field of organic polariton condensates. The candidate is co-author in three papers, two related to the dissertation's topic, one as first and one as second author. Although the first author paper is still under review, the overall publication list, and the quality of the obtained results, is sufficient for a PhD.

Apart from the aforementioned problem with the English presentation there are a number of minor points I wish the candidate to address before the viva:

- 1) I cannot find the definition of ϵ in Eq. 2.7.
- 2) On page 27 Ω_0 should be called Vacuum Rabi Frequency, to distinguish it from the AC-Stark shift case.
- 3) On page 27 it is said that strong coupling is realized if the coupling is **much** larger than the dissipation. The candidate should define what much means in this context.
- 4) What are the features appearing on the corners of Fig. 2.5?
- 5) In section 2.2.1 it is stated that condensation and Bose-Einstein condensation are not the same thing. This should be properly discussed.
- 6) On page 38 ASE should be properly explained.
- 7) Through the manuscript there are a number of equations not provided in symbolic terms (e.g., beginning of page 39). This is incorrect. Both the quantities on the left and right hand sides of every equation have to be provided also in symbolic form.
- 8) On page 43 there is a meV missing.
- 9) Section 3.4.2 is difficult to understand. It should be rephrased and better explained.
- 10) What is the black polygon in Fig. 3.10?
- 11) In Fig 3.10 units are missing from the top right corner.
- 12) On page 53 a procedure to estimate the threshold incident excitation density is discussed but it is not clear to me whether the data are presented. If they aren't please add them.
- 13) In section 3.4.4 I could not find the definition of "d".
- 14) In Fig. 4.9 it is not clear to me how the data points in panel a) are reduced to these in the other panels.

- 15) This is just a comment on which the candidate should not feel obliged to act, but why between panels b) and c) of Fig. 4.9 the same energy is referred once as 120meV and the other as 0.12eV?
- 16) Fig. 4.10 definitely rules out a quadratic dependency, but leaves some space for a linear one. As such the discussion about the clamping of the excitation density presented on page 72 should be expanded and clarified.
- 17) The origin of the square root dependence of the vacuum Rabi frequency upon the number of molecules should be discussed.
- 18) Fig. 4.14 clearly shows how the steepness of the blueshift increases with the intermolecular energy transfer. Given that all the results of chapter 4 rest on this fact, a better microscopic explanation would be appreciated.
- 19) I am not sure I understand the first equation on page 79. On page 78 F_c is defined by the equation $N_0^{\parallel} = N_0^{\perp} = F_c N_{\text{tot}}$. N_0^x is the number of out-of-plane molecules, but shouldn't it include also **all** the uncoupled molecules? If that's the case $N_0^x = N_{\text{tot}}(1 - 2F_c)$. Otherwise what is the difference between a molecule which is uncoupled because its dipole is orthogonal from one whose dipole has some other form of disorder? Orientational disorder has a role similar to other forms of disorder, as shown for example in PRB **71**, 115320 (2005).

Provisional Recommendation

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

I recommend that the candidate should defend the thesis by means of a formal thesis defense only after appropriate changes would be introduced in candidate's thesis according to the recommendations of the present report

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