Skoltech
Skolkovo Institute of Science and Technology

# Thesis Changes Log

**Name of Candidate:** Baryshev Stepan

**PhD Program:** Physics

**Title of Thesis:** Photon correlations of optically trapped polariton condensate

**Supervisor:** Pavlos Lagoudakis

**Co-Supervisor:** Anton Zasedatelev

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

Dear Jury, in order to satisfy your comments, the following changes where done to the thesis.

Corrections to the comments from **Prof. Simone De Liberato**:

Comment: I would suggest the Candidate to develop better these (the spinorial structure of the polaritons or the concept of Stokes vector) concepts in this introductory Chapter in order to make the thesis more broadly accessible.

Answer: an additional comment was added to the thesis explaining the concepts, "They can undergo power-driven Bose–Einstein condensation [Bose–Einstein condensation of exciton polaritons, J. Kasprzak, et.al., Nature volume 443, pages409–414 (2006)] into a spinor order parameter corresponding to the right-hand and left-hand circular polarization of the emitted light like, in a conventional semiconductor spin-laser. The complex order parameter cannot be directly observed in a photoluminescence (PL) experiment, however resolving PL in polarization bases gives access to the components of the condensate psuedospin (Stokes) vector [Stochastic polarization formation in exciton-polariton Bose-Einstein condensates, D. Read et.al., Phys. Rev. B 80, 195309], providing the orientation and the degree of of polarization, which will be discussed in Section 3.2.3."

Comment: I would suggest to develop a bit more this part (conclusion) with some more personal considerations.

Answer: the conclusion section was expanded accordingly, as well as conclusion sections were added in the end of chapters 2, 4 and 5.

Comment: the quality of the English could be improved, as at times it makes the manuscript difficult to understand.

Answer: the thesis went through thorough grammar and semantic check and errors were fixed.

Comment: in the Abstract the transition between Chapter 2 and 3 is too abrupt. The Abstract needs to provide a narrative for the whole manuscript, not piecewise descriptions of the different Chapters.

Answer: the abstract was reworked to have a smoother transition and better connect the work done for different systems.

Comment: in the Introduction all even-numbered Chapters are “the last Chapter”.

Answer: this error was fixed in the renewed version of the thesis.

Comment: in the Introduction concepts as “HOM dip” or V are used without being defined. Being this the Introduction, a formal definition is not needed, but the reader cannot be expected to know them already.

Answer: the introduction chapter was reworked accordingly.

Comment: the description of IRF, SER, and their relation in TCSPC in Section 2.1 is not clear and has to be expanded.

Answer: additional information was added to the according section, to explain the abbreviations and their physical meaning. The added text begins with “The IRF reflects how a infinitely short signal is seen by the instrument, and SER is the actual detector output pulse for a single photoelectron created through the process of effective photon absorption and amplification through an avalanche process within the detector.”

Comment: in Section 2.2 the origin of the dependence of the work function upon the spheres dimension has to be explained.

Answer: the appropriate passage was added starting with: “The mentioned work function is formally known as an energy required to remove an electron from a solid to a point in the vacuum immediately outside the solid surface.”

Comment: in Section 2.3 it is not clear how the peaks are correlated with specific chain lengths.

Answer: to add information on this matter the following sentence with appropriate citation was added:
“The energy gap between the highest occupied molecular orbit (HOMO) and the lowers unoccupied molecular orbit (LUMO) decreases with chain length and shows a trend toward a finite gap of 0.48 eV with increasing length of the carbon chains”

Comment: equation 3.10 is inverted.

Answer: the equation for exciton creation and annihilation field operators, as well as the last part of the section 3.1.3, addressed by the Jury in the next comment, was removed from the thesis text due to lack of the necessity for the explanation of this work.

Comment: in Section 3.1.3 the last sentence about the Mott transition should be qualified. The Mott transition is due to saturation, not Coulomb interaction.

Answer: the last part of the section 3.1.3 was removed from the thesis text due to lack of the necessity for the explanation of this work.

Comment: in Section 3.2.1 the sentence about “reducing the phonon density of states” should be better explained.

Answer: this passage was reworked to be more clear.

Comment: in Section 3.2.1 the sentence stating “reaching thermal equilibrium is hardly possible” should be qualified, as there are publications claiming to have done that.

Answer: the statement was clarified and wording was changed. The according citation, claiming reaching thermal equilibrium in high Q-factor micro cavity with optically trapped polaritons in big trap [Bose-Einstein Condensation of Long-Lifetime Polaritons in Thermal Equilibrium
Y. Sun et.al., Phys. Rev. Lett. 118, 016602] was added.

Comment: in Section 3.2.3 the spinor nature of the polaritons, the concept of pseudospin, and the concept of Stokes vector and Poincaré sphere have to be clearly introduced.

Answer: the section was reworked to facilitate the necessary concept more clearly. Starting with "Polarization of light can be described using a set of values $S_0$ , $S_1$ , $S_2$ and $S_3$ called Stokes parameters."

Comment: the quantities in Eq. 3.20 are not defined.

Answer: the definition for the Stokes vector, the Stokes parameters and how they are measured were added to the renewed version of the thesis. As well as the definitions for the corresponding measured intensities.

Comment: in Section 4.1.1 the parameter eta is used before being defined.

Answer: this is fixed in the renewed version. Parameter η, denoting the quantum efficiency of the detector, is explained right after it is introduced.

Comment: in Section 4.1.1 the statement about the time-spatial domain D and its relation to the discussion has to be better explained.

Answer: the described domain **D** bears a technical meaning of domain within which the measuring apparatus perform to obtain the correlation functions. Additional clarifying information was added to the thesis, "Here **D** is the domain of integration for the total energy collected by the detector an lies within the boundaries of **t** ∈[0,T], **r** ∈A and bears technical limitations of measuring apparatus"

Comment: in Section 4.2 the concept of polariton blockade has to be defined.

Answer: the following explanation was added to the renewed thesis
"As authors claim, if the interaction-induced energy shift is larger than the polariton linewidth withing the highly confined potential, then the system is said to be in the polariton blockade regime, manifesting itself as photon anti-bunching effect, lowering of $g^{(2)}(\tau)$ function values for delays $\tau$ smaller than the polariton lifetime."

Comment: in Section 4.2.3 the link between the reservoir and the out-of-plane magnetic field could be more explicitly explained.

Answer: a following sentence was added. "Excitation of the reservoirs in the unbalanced fashion, with elliptically polarized pumping, provides more gain to one of the spins, and effectively splits the energy of the system for $\sigma^{(\pm)}$, as if there is an effective magnetic field present."

Comment: the choice of parameters in Section 4.2.5 should be justified.

Answer: the exact parameters values discussed in this section were derived for our sample to nicely match the theory with the experiment. The values were used to explain the experiment of our previous works, where they describe the observables well enough. To clarify this part, the following text was added to the thesis, "This set of parameters are of the typical values for our sample, and has been used for our previous works in which they nicely matched the theoretical predictions with the observed experimental results."

Comment: in Section 4.2.5 the reason Theta should be understood as a quarter waveplate should be more explicitly explained.

Answer: to clarify this part a following addition to the thesis was made in corresponding section, "Theta can be understood as a quarter waveplate angle in the experimental setup which determines the polarization of the incident light, since depending on the QWP orientation the polarization ellipticity of the excitation beam can be tuned, effectively changing the pumping power for + or - reservoirs,"

Comment: in Chapter 5 what is plotted in the graphs has to be properly defined and explained. “HOM dip”, V, Coincindence, have all to be defined in the main text in terms of physical observables and clearly referenced in all the images. All colorplots need a colorbar, and all colorbars need a label.

Answer: all the above mentioned errors were fixed accordingly.

Corrections to the comments from **Prof. Daniele Sanvitto**:

Comment: yet the description of the dynamics of the excited carriers are well understood and backed up by a theoretical analysis–although this is only present in some of the publications of the candidate but missing in this thesis.

Answer: a new figure was added in the end of Chapter 2 to qualitatively show the theoretical and experimental matching of the carrier dynamics in chains of different length. The text referring to the figure was, also, added, and coherently introduced to the chapter.

Comment: the grammar though should be improved.

Answer: the thesis went through the thorough grammar and semantic check and the errors were fixed.

Comment: I did not find the last chapter in any publication, however it is possibly in preparation.

Answer: the publication correspondent for the results of this chapter is indeed under preparation at this time. The sentence explaining this was added to the Conclusion section of Chapter 5 stating:
“The theoretical framework as well as publications regarding these new observations of HOM effect in trapped polariton condensates is under preparation at the moment.”

Comment: I also suggest checking the consistency of the phrases and figures. For instance, references to figures 4-3, 4-4 and 4-5, 4-6 in chapter 4 or the labels which sometimes are too small, like those at the end of chapter 5.

Answer: the issues with the references to the figures were checked and fixed. The figures in Chapter 5 were reworked to better represent the results, and have higher readability.

Comment: also it would be nice if some concepts like “the condition of tilted limit cycle regime” would be explained rather than given for granted.

Answer: to address the comment a following corrections were made for the thesis text in Chapter 3, section “Polarization of polariton condensate”:
“For the elliptically polarized excitation, the condition of tilted limit cycle regime can be fulfilled. This effect can be viewed as periodic precession of the condensate pseudospin around some effective magnetic field, emergent due to structural and pump induced energy splittings in the system, discussed in more detail in Chapter 4, which manifests itself as an effective depolarization of the condensate emitted light, when measured with a time resolution less then the cycle period.”

Corrections to the comments from **Prof. Dmitry Gorin**:

Comment: Page 17, please write nano- and particle as one word – nanoparticles.

Answer: the thesis went through thorough check and grammar mistakes were fixed. Including this one.

Comment: Page 17, Adding DLS results as well as TEM images of gold nanoparticles could be useful.

Answer: the process of the sample fabrication is a complicated process and not the subject of this dissertation. However, an correction was added to the thesis, guiding a reader to the detailed information on dynamic light scattering technique applied and TEM images of carbon chain with golden nanoparticles with the following sentence:

"The fabrication process of carbon monochain samples is a sophisticated process and not the main topic of this thesis, thus only few details are given here. However, thorough explanations are given in the supplementary material of Ref {doi:10.1021/acs.nanolett.0c02244} where information on dynamical light scattering and carbon chains with visibly attached gold nanoparticle TEM images are presented."

Comment: I did not find any information about substances that have been used for gold nanoparticle surface modification as well as description of gold nanoparticle synthesis. This information should be added to the PhD thesis.

Answer: our collaborators chemists provided the samples, and unfortunately, I am not aware of many details of sample fabrication process at this point. I am aware that there are methods of fabrication pioneered by J. Turkevich, et.al. [A study of the nucleation and growth processes in the synthesis of colloidal gold, Discuss. Faraday Soc., 1951,11, 55-75] and G. Frens [Controlled Nucleation for the Regulation of the Particle Size in Monodisperse Gold Suspensions, Nature Physical Science volume 241, pages20–22 (1973)], which are capable of providing gold nanoparticles of sizes ranging from 10 to 100nm. The appropriate citations were added to the thesis.

Comment: why did author use the gold nanoparticles at two sizes 10 and 100 nm? Please add an explanation of this chose as well as errors for nanoparticle size evaluation;

Answer: the reasoning behind having the nanoparticles of different sizes is explained with addition of the appropriate passage starting with:
"The mentioned work function is formally known as an energy required to remove an electron from a solid to a point in the vacuum immediately outside the solid surface." The explanation of nanoparticle size evaluation is explained in the added sentence: "The size of the nanoparticles were estimated by TEM images and near infrared transparency spectra (not shown)."

Comment: Page 18, Figure 2-2 (b), why do not I see any gold nanoparticles in the HR-TEM images?

Answer: the following sentence was added, explaining this, "Gold nanoparticles remained outside the frame of the image in this figure."

Comment: I think that results of measurements of the extinction spectra of gold nanoparticle/carbon chains could be useful.

Answer: unfortunately the extinction spectra where not measured during the optical investigation of these samples and I am not able to provide it at this point.

Corrections to the comments from **Prof. Mailis Sakellaris**:

Comment: I found that the thesis has a number of minor typos the corrections of which would improve the clarity of the text.

Answer: the thesis went through thorough check after which grammar and semantic errors were fixed.

Comment: furthermore it is my opinion that the abstract is too specific and technical and lacks the vision and motivation elements.

Answer: motivation, as well as vision, are described in reworked introduction and conclusion sections. The abstract of this work is meant for a reader get an idea of the achieved scientific results and methods used. The abstract section was reworked to better facilitate the transitions between the three major parts of the thesis work.

Comment: likewise I found the conclusions/outlook section is too brief.

Answer: The conclusion section had minor revision, as well as additional conclusion sections were added at the end of each chapter.