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Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Aly Mohamed Tawfik Aly Elakshar

PhD Program: Physics

Title of Thesis: Single-walled carbon nanotubes in top cell for perovskite-silicon tandems

Supervisor: Professor Albert Nasibulin

Name of the Reviewer: Anatoly Pushkarev

I confirm the absence of any conflict of interest (Alternatively, Reviewer can formulate a possible conflict)	**Date: 19-11-2024**

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

This thesis by Aly Mohamed Tawfik Aly Elakshar is devoted to modern optoelectronic devices for photoelectric conversion. The Author employs single-walled carbon nanotubes (SWCNTs) film as an electrode in organic-inorganic lead halide perovskite top cell combined with a silicon bottom cell. The motivation of engineering the complex four terminal tandem solar cells is explained by overcoming Shockley-Queisser limit. The choice of SWCNTs film is justified by its high transparency, low resistivity, flexibility and last but not the least the inertness of carbon materials to chemical reaction with halide perovskites. Perovskite cells with and w/o hole transport layer (HTL) are fabricated. Phenomenon of charge carrier extraction is studied in detail by means of time-resolved photoluminescence (TRPL) spectroscopy technique. An original strategy of passivation surface defect sites at perovskite grain boundaries in polycrystalline films is employed. The Author shows the deposition of methylammonium iodide not only converts the excess of lead iodide to perovskite but also enhances PL intensity due to the reduction of number of nonradiative electron-hole recombination channels. Besides basic characterization of solar cells efficiency and other crucial parameters, impedance spectroscopy (IS) of perovskite cells is conducted and fair conclusions about charge transfer at perovskite/SWCNTs and HTL/ SWCNTs are made based on IS data. Finally, a tandem cell exhibiting 24.34% power conversion efficiency is demonstrated.

In my opinion, the quality of the thesis is excellent. It is well-written and has a good logical structure. The whole story begins from the strong motivation, then basic characteristics of solar cells are presented, perovskite materials structure and properties are described, Shockley-Queisser limit definition and strategies to overcome it discussed. This brings a reader to the main object – tandem solar cell – smoothly. Thereafter methods and materials are described followed by device fabrication and characterization sections. In the end, the Author makes his conclusions and short notice about perspectives for perovskite-silicon tandems with SWCNTs electrode.

The topic of the thesis is certainly relevant to its actual content. The experimental methods section has a list of commonly used procedures recognized by the scientific community as sufficient for obtaining reliable and reproducible results.

The scientific significance of the results is major. The Author declares the device maximum efficiency 24.34%. State-of-the-art perovskite-silicon tandem cells produced in many groups around the globe show similar efficiency. Although, their efficiency is comparable with that of counterparts fabricated by Author, the latter, indeed, possess a serious advantage over the former ones since carbon electrode does not react with halide perovskite. Therefore, perovskite devices with carbon electrode show much longer operational stability as compared to ones with metal electrodes. Quality of publications is high and corresponds to the international level.

During the reading of thesis, I figured out some points that require clarification. Although, they do not lower down the significance of the obtained results, I encourage the Author to address the following comments:

i) A complete expression for J-V curve of solar cells contains series and shunt resistance. How large these values in the fabricated top cells are? What is their impact on photoelectric conversion efficiency?

ii) The Author uses Tauc plot approximation of the optical absorption spectrum edge to derive a band gap energy value (E_g). This approximation is applicable to square root relation between absorption and photon energy since optical transitions in a direct semiconductor are supposed to occur between two parabolic potentials in the conduction and valence bands, respectively. In the case of halide perovskites, the absorption spectrum edge has a complex lineshape. It consists of band-to-band and excitonic absorption. The latter one could be represented as a sum of Lorentzian peaks corresponding to optical transitions from the ground state to discreet excitonic levels and cannot be approximated by Tauc plot. Therefore, I think there is a small error in the derived value E_g. Could the Author comment about the accurate estimation of E_g and evaluate the error?

iii) Looking at equation for charge carrier dynamics one can see ABC model reported by Shen et al. [Appl. Phys. Lett., 2007, 91, 141101] is adopted in the thesis. In this model charge carriers do not reside in trap states and undergo rapid nonradiative relaxation. As a results, the concentration of excited electrons (n) and holes (p) is the same and can be described by n solely. However, the Author introduces p, n, and their lifetime τ_p and τ_n, that seems to be important when Shockley-Read-Hall model [Phys. Rev., 1952, 87, 835; Phys. Rev., 1952, 87, 387–387] is considered for the description of charge carrier dynamics. I was wondering if there is a particular reason for the mixing two different formalisms?

iv) I noticed bi-exponential approximation of PL decay in Figure 3.3. If the Author employs ABC model and neglects third order member k_3n^3 associated with Auger nonradiative recombination, the solution of the equation (18) cannot be bi-exponential function. The Author should explain his approach to calculation of lifetime values for bi-molecular and monomolecular decay.

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*