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Review of doctoral thesis:

"Transient optical and electrical phenomena in organic semiconductors: Insights into fundamental photophysics, resistive switching, and emulation of synaptic plasticity"

submitted by Yadu Ram Panthi

The dissertation thesis submitted by Yadu Ram Panthi focuses on unraveling transient optical and electrical phenomena in organic semiconducting materials. The thesis strives to provide insights into the fundamental physics and resistive switching properties paving the way towards electronic memory and synapse-mimicking functionalities.

The thesis consists of abstract, introduction, materials and methods section, results and discussion parts summarizing the obtained main results and conclusions section. The submitted thesis has 103 pages and references the 3 main publications (all published as first author) and 1 Czech utility model referenced as appendixes from which it mainly sources. The work is well concepted and organized with a clear presentation and vision. The introduction provides a careful resume on the physical phenomena and prerequisites for the development of optoelectronic devices. Furthermore, it addresses the operation mechanisms of the envisaged electronic devices. However, I would have appreciated if the introduction addressed in a greater detail the actual gap of knowledge within the field. Nevertheless, this has been amended to certain extent within the individual Result and Discussion sections. The main findings are elaborated within three subsections of:

1) Photo-physics of TDPP derivatives: Insights into ultrafast processes;
2) Organic memristors: Electronic memory and emulation of synaptic plasticity;
3) Role of dipolar polarization and interface optimization on the performance of OFETs.

The candidate addresses and explains all obtained results independently going well beyond his published works. Despite the fact that the work is rather precisely written, I have remark that it lacks precise definition of the aims of the thesis in a separate section. Despite the fact that these can be inferred from the Introduction and the individual Results and Discussion sections, an independent Aims sections would add towards more explicit structuring of the thesis. The Conclusion section is written as a general conclusion stemming from the work of the team and might have been better to be addressed from the point of the Author.

Despite that the thesis is well written and well readable, I have to point out few discrepancies, typos and inconsistencies:

i) missing captions of TOC images presented at the individual chapters;
ii) stylistic alignment issues at the end of individual pages (example end of Page 2, 3, 6, 7, etc.);
iii) the Author did not use cursive for symbols of physical variables (equation on Page 9, equations of Page 12, Page 71 etc. to name just a few);
iv) inconsistent use of dash and hyphen symbols (Page 40);
v) typos on physical units (Page 54 photon.pulse^{-1}.cm^{-3});
vi) smaller font size/or different style on Page 62 on write-read-erase-rereader;
vii) incomplete List of abbreviations;
viii) questionable usage of SEC abbreviation (does it refer to Size-Exclusion Chromatography?).

However, the pointed-out discrepancies, typos and inconsistencies are minor and do not seriously affect the quality of the dissertation. As previously noted, the thesis mainly builds on 3 publications (all of which published as first author) of a high scientific visibility. The included utility model proves the applicability of the results achieved in this thesis. In my opinion the applicant has shown a high level of

independence, diligence and creativity, thus undoubtedly demonstrated to be fully capable of performing independent research. As to date when this review is being submitted the applicant has(co-)authored 7 publications and contributed to international conferences through poster and oral presentations.
At the end I would like to express my agreement with the Applicant's concluding remark, that **all these results achieved within this thesis contributed to the understanding of the physical nature of the studied phenomena in organic materials and can be further exploited in materials science and device engineering in organic electronics.**
Therefore, I **recommend Yadu Ram Panthi´s dissertation for defense at the P4F4 Chair of Biophysics, Chemical and Macromolecular Physics, Faculty of Mathematics and Physics, Charles University.**

Specific questions to the dissertation:

1) The candidate elaborates the TUV-vis optical absorption and fluorescence emission spectra of PCaPMA and PVCa films. Can he further elaborate them? What was the molar mass of used polymers? What was the film thickness of the individual films? (Page 58-59, Figure 3.6)
2) The candidate states "The HOMO levels of PCaPMA and PVCa were determined to be -5.23 and -5.34 eV, respectively. In these polymers, the LUMO positions could not be measured electrochemically, they were derived from the respective HOMO positions and the bandgap obtained from the Tauc plot of the optical absorption spectra. Using this approach, the LUMO positions of -1.72 eV and -1.81 eV, were obtained for PCaPMA and PVCa, respectively." (Page 61).
   Can the Author elaborate the procedure in a greater detail? What are the uncertainties of determined HOMO and LUMO values? Are they significant to claim the differences between the two systems?
3) Figure. 3.11 presents the Schematic of the Pavlonian learning algorithm and graphically relates it to obtained results within the same panel. However, it seems to me that the relation deserves further explanation. Can the applicant elaborate the connection to a greater extent beyond the impression created by the image? (Page 65)
4) The Author states: These results underscore the potential of these devices to emulate memory processes and mimicking learning, required for AI development. (Page 65).
   Can he further explain the interconnection between ITO | PCaPMA | Au device and AI development?
5) Within Sub-section 3.3, the Author utilizes trimethoxy(2-phenylethyl)silane (PETMS) for the creation of a SAM though a silanization? While the applicant supposes the assembly of a monolayer the used silane is prone to polymerization and formation of a siloxane multilayer. Can the Author comment on the thickness of the PETMS?
6) Does the Author have any experimental/spectroscopic proof on the vertical structuring of the various multilayer sandwiched structures presented within the thesis?

In Prague, 31.08. 2024

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