



## Report on the Ph.D. thesis by David Paleček (Quantum Coherence for Light Harvesting)

The dissertation thesis by David Paleček is based on eight papers, of which three were published in high-ranked journals (including one in the Nature Group), and five are reported as manuscripts. For five of these papers or manuscripts, David Paleček is the first author with the dominant contribution to both experimental and theoretical parts of the papers. The thesis is of high quality, both from the content and presentation point of views. The content of the thesis demonstrates authors' expertise in various fields extending from theory of nonlinear spectroscopy to knowledge of various photosynthetic systems.

The thesis is very coherent in a way that it clearly focuses on one particular problem (long-lived coherences in artificial and natural light-harvesting systems), which is tackled by a specific spectroscopic method (2D electronic spectroscopy). The author faced a real challenge during his work on this thesis; first, the problem of coherences in photosynthetic light-harvesting systems is a hot topic of current science that remained unexplained and second, he has decided to go a bit against the main stream, which is always extremely difficult. However, the presented thesis proves that the authors succeeded in both aspects. The thesis provided feasible explanation of the long-lived coherences, which are explained as of vibrational origin, in contrast to the 'fancier' electronic origin often promoted in the literature during the past years. I also like an imprint of development of ideas in the thesis as the first paper still retains the long-lived coherences of electronic origin, while in later papers this turns into vibrational origin in the ground state (Paper IV and later).

The thesis has a very nice introductory part consisting of chapters devoted to overview of studied systems (Chapter 2) and used methods (Chapter 3). The introduction is very well written and serves precisely the purpose that a good Ph.D. thesis should do – it is a great reference material for potential successors in the field, containing many details concerning both the theory and experiment that the reader will not find in a 'standard' scientific paper. Any new Ph.D. student or postdoc will greatly benefit from reading the introduction of this thesis as the author slowly guides the reader through various aspects of the experiment, theory and the systems he has studied.

The key results achieved during the work on this thesis are summarized in Chapter 4, in which the author describe in details development and principles of a newly-introduced photophysical process, the Energy Transfer Induced Coherence Shift (ETICS). I consider this part, together with Paper IV, as the core of the thesis and also as a great achievement of David Paleček's efforts during his Ph.D. project. It is quite rare to see a proposal of a completely new photophysical mechanism in a Ph.D. thesis; it underscores the outstanding quality of the whole thesis.

All papers (except Paper II that somehow does not fit into the whole story) included in the thesis deals with long-lived coherences in light-harvesting systems and demonstrate the power of the 2DES method, especially when in hand of a devoted specialist. The Paper I defines the long-lived coherences in the reaction center of purple bacterium *Rb. sphaeroides*, a system that is used thorough the whole thesis as a model system. The results of Paper I, obviously taken during the initial stages of David Paleček's Ph.D. adventure, most likely formed the further direction of the research presented in the thesis. I can only guess, but I assume that neither the author nor his supervisor were satisfied with the explanation of the long-lived coherences




presented in Paper I and decided to tackle this problem in more detail. This led to the proposal of ETICS introduced in Paper IV and further developed in Papers VI and VIII. The collection of papers is complemented by two rather technical studies (Papers V and VI) which both deal with technical aspects of a 2DES experiment and suggest some improvements for future experiments. This again demonstrates the broad range of knowledge of the author who proved his ability to deal both with theoretical and experimental aspects of 2DES.

To summarize, the whole thesis is clearly written in a very good English, with very nice introduction understandable even for a non-specialist on one hand, and cutting edge science in the last chapters on the other hand. This thesis falls into the top 10% of Ph.D. theses I have seen so far. One may have some reservations to the fact that the key results are presented only as manuscripts in the thesis, but given the shift of the paradigm proposed in the thesis, it is understandable that it has been a long journey to the final results.

Overall, I have no reservations that this thesis fulfills the requirements of the PhD work in any country or any university. The candidate did a great piece of scientific work and proved that he is able to contribute significantly to the development in the field. The thesis in my opinion fulfills (and in many aspects exceeds) the criteria for being accepted as a ground for awarding David Paleček with the title PhD. and should be accepted for the defense.

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