

Supervisor's report

on doctoral thesis entitled

Ultrafast response of electrons in nanostructured and disordered semiconductor systems studied by time-resolved terahertz spectroscopy

prepared by **Mgr. Vít Zajac**

This thesis is focused on an investigation of ultrafast dynamics and transport of photoexcited charge carriers in semiconductor nanostructures. In the literature, the time-resolved terahertz conductivity spectra were frequently interpreted in the framework of phenomenological theories (e.g. Drude-Smith model). In some works the localized plasmon resonance (which can be described equivalently by effective medium theory) was used to interpret the conductivity behavior in larger nanostructures or in microstructures. More recently, Monte-Carlo calculations were introduced to describe the local charge carrier motion inside nanocrystals. However, a more general approach, connecting the local charge carrier transport and the build-up of depolarization fields in the nanostructures, was lacking. The present work sets up this connection. It develops a theoretical framework for the evaluation and interpretation of ultrafast terahertz photoconductivity phenomena and applies it to several particular systems (silicon nanocrystals, titanium dioxide); in my opinion it constitutes a significant contribution in this rapidly growing research field.

Referring to more than 130 bibliographical references the author covered the current state-of-the-art of the optical pump — terahertz probe spectroscopy and described the most important semiconductor systems to which it has been applied so far. He provides a survey of the experimental work and various interpretation frameworks that have been used in the literature.

The thesis probably does not represent a very easy reading, especially for a non-specialist in the field. However, interpretation of the THz spectra on a microscopic level and disentangling various phenomena contributing to the spectra is not an easy task in any way and requires a complex formalism. Keeping this in mind, I believe that Mr. Zajac has chosen the appropriate approach in the redaction of the manuscript, which permitted him to elaborate a scientifically rigorous and understandable work.

The author shows that, in order to understand the nature of the microscopic behavior of carriers well beyond the phenomenological fitting, various complex phenomena must be carefully described. Based on electrostatic simulations he suggests an effective medium model parametrized by three morphological parameters; this model is able to describe both non-percolated samples and samples with complex percolation pathways. On top of this model he solves the wave equation for the THz wave in inhomogeneous (percolated or non-percolated) photoconductive medium. The solutions provide equations, which connect the THz transient transmission data to the microscopic mobility spectra of carriers and to the sample morphology.

Experiments in various selected systems were performed very carefully: generally as a function of temperature and as a function of the optical pump pulse fluence (i.e. initial carrier concentration). The initial pump fluence was varied typically over three orders of magnitude leading to very long data averaging process during acquisition (12h or more) at low pump fluences. However, this approach is required for truly microscopic interpretation. In addition to particular information about the studied samples the author obtained some results with a quite general validity concerning the response of non-percolated samples and concerning the importance of the size distribution (and uncertainties in the size distribution) of nanocrystals

in the sample. Besides the main focus of the thesis, Mr. Zajac also participated with enthusiasm to other developments of the laboratory facilities; for example, he designed a primary vacuum chamber, which is now used as a principal environment for steady-state THz measurements.

Mr. Zajac mastered very well the terahertz time resolved experiments. As I already mentioned, his work style is quite rigorous. He always thoroughly thought out the physical implications of various models (which were quite complex in some cases), tested them and fitted their numerous simplified variants to the data to be completely sure about the plausibility of his interpretations. The only criticism that I may express in his regard concerns the quite lengthy process of the thesis manuscript preparation. Surely, the manuscript could have been written faster; on the other hand, with the time the manuscript matured to a very well thought and self-consistent form.

During the doctoral studies Mr. Zajac has shown his skill to perform very good quality scientific work and also his ability to present the results on an international level (4 publications in impacted journal, 2 oral contributions and 5 posters at international conferences, successful participation at the Fame Lab 2013 competition on a national level). The thesis presents original results and I strongly recommend accepting it for the defence.

In Prague, March 24, 2017

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