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Opponent report

for the doctoral thesis

“Ultrafast photoconductivity and charge carrier transport in semiconductor nanostructures: a study by terahertz spectroscopy”

Submitted by **Mr. Vladimir Pushkarev**

at the Faculty of Mathematics and Physics (Charles University)

The thesis reports on an in-depth study of electronic transport properties in semiconductor nanostructures investigated in a contact-free manner by THz time-domain spectroscopy, more precisely optical-pump terahertz probe experiments. Semiconductor nanostructures are a vital field of interest as their physical properties can be tailored by controlling size, composition and arrangement. With many ways of top-down and bottom-up fabrication they can be furthermore produced for nanoelectrnic device applications. Knowledge and understanding of the transport properties is of key importance both for fundamental studies and device applications. With shrinking size, ballistic transport in mesoscopic structures (mean free path larger than dimension) and confinement in quantum structures (dimensions on the scale of de Broglie wavelength) become important. Furthermore, surface and interface effects such as surface fields and edge states play an important role. THz time-domain spectroscopy is known to be a powerful method to study transport properties without the effects of electrical contacts. However, most previous studies on inhomogeneous materials with carrier confinement have been analyzed with the phenomenological Drude-Smith model that can provide only limited insights and no real physical understanding. The author has used a number THz spectroscopic techniques and combined them with advanced physical models developed in his research group. The resulting thesis is a substantial advancement in the field. It leads to physical understanding of electronic transport properties in controlled nanostructures and paves the way to analyzing complex systems where many effects contribute to features in the THz spectra.

In the introduction, the author explains the difficulties of studying local transport properties of inhomogeneous materials. He provides an overview of available methods and briefly describes the types of nanostructures used for his studies. The introduction ends with a brief overview over the chapters of the thesis.

In the first chapter, various techniques of THz time-domain spectroscopy are introduced. The author describes their principle, capability and limitations regarding spectral range and the very basics of obtaining spectral information in amplitude and phase of a sample. The introduced techniques are standard optical pump – THz probe (OPTP) spectroscopy, multi-THz OPTP spectroscopy using air plasma generation and air-biased coherent detection, and near-field THz spectroscopy.

Chapter 2 discusses the THz response of photoexcited nanomaterials. Starting from Maxwell’ equations the wave equation is derived and the general relation between pump-induced dynamic conductivity and differential transmission (and reflection) is provided. Then different models for the description of the THz conductivity of semiconductor nanostructures are presented. Furthermore it is described how the modelling results are employed into the wave equation in order to fit experimental spectroscopic results.

In Chapter 3 the experimental setups for the techniques introduced in Chapter 1 are presented with all technical details.

Chapter 4 and 5 contain the experimental results, their analysis and the conclusions. Chapter 4 is focused on the investigation of Si nanocrystals embedded in a SiO_2 matrix. For better size control a superlattice approach is pursued. Analysis of the OPTP spectroscopic data, in particular at low temperature, requires model including quantum confinement. At low THz frequencies, tails from the resonant peaks for different size contributions are seen. A refined model, that takes into account the thermalization current properly, is presented after the analysis with an initially slightly simpler model. The main conclusions remain the same but there are quantitative differences in the contributions assigned to different nanocrystal sizes. Next, multi-THz spectroscopy is successfully applied. This seems very promising to obtain more direct information on the conductivity of quantum confined electrons in the nanoparticles as the resonant peak fall into the accessible spectral range. However, as phononic resonances of SiO_2 and layer thickness effects play an important role, the analysis is rather complicated. The author thoroughly sorts the influence of various effects and by that is able corroborate the results on the nanocrystal response obtained at lower THz frequencies, while the inhomogeneity of the SiO_2 regarding composition and strain does not allow firm conclusions on its exact dispersion.

In the last chapter spectroscopy on a regular arrangement of single-crystalline III-V nanowires is performed. The author mentions an initial approach using InGaAs nanowires on InP substrate. Here, however, it turned out that the substrate contribution dominates the spectroscopic signatures. Then a more demanding process to transfer GaAs nanowires to a sapphire substrate is described and large area regular arrangements are achieved even though the wires are just van der Waals bonded to the substrate. From these structures very clear spectra of quantum-confined electrons are obtained. The analysis shows that the carriers are confined in smaller spatial regions than expected from the sample geometry. The author convincingly explains the discrepancy by surface field effects. This is corroborated by near field microscopic studies paying attention to the signal content of the harmonic contributions from the cantilever oscillation that provide qualitative depth information. First attempts to quantify this in a simple model of dipolar response are presented.

The thesis is summarized with a conclusion and a detailed description of the personal contribution of author in the different tasks of the whole project.

The work of the author is highly original, the experimental skills and thorough work have provided an excellent sample (the regular arrangement of monocrystalline GaAs nanowires) and THz spectroscopic datasets of high quality. The author applies complex model with great care and self-critically judges his own results in particular when many fitting parameters are involved. The thesis is organized in a clear manner and is well written, the balance between text, equation and figures is very good. There are some very minor issues that could be improved, for example in the distribution of information in Chapter 1 and 3 and by showing some didactic figures indicating where a particular model or fitting approach does NOT give good agreement with experimental data to highlight clearly why the finally chosen path was the most reasonably one.

I have the following questions to the candidate:

1) If I compare Figure 28 and 30 (Page 52 and 55, respectively) the increase of the curves towards higher frequencies (in particular for $x = 0.3$ photon fluence 7.8×10^{12} cm^{-2}) in differes quite a bit for the simpler and the more refined model, but for the particular curve the simpler model captures the trend better. Can the author comment on this? Can it be that the refined model under certain conditions overestimates effects related to the thermalization current?
2) On page 72 it is mentioned that the $In_{0.53}Ga_{0.47}As$ / InP was chosen, because it is commercially available. Can the author explain, why this particular composition (53 % In) of InGaAs is commercially available on InP substrates?
3) Finally a more general question regarding THz technology: On Page 4 the author correctly mentions that THz quantum cascade lasers (QCLs) require cryogenic cooling. Why is this not the case for the nonlinear Thz generation in ZnTe used in this thesis. To go one step further: In 1970 THz gas lasers (often methanol lasers pumped with a CO_2 laser) were invented. They are bulky and not extremely user friendly, but they lase at room temperature. What is the reason for this difference compared to THz QCLs?

Overall this thesis is of very high quality, I cannot find any signs of scientific misconduct in presented work. The author has shown his ability to perform an independent research work. I recommend the doctoral thesis for the defense in order to author can be appointed as a candidate for a PhD title.

Dr. Stephan Winnerl   Dresden, 28.02.2023