Skoltech
Skolkovo Institute of Science and Technology

Thesis Changes Log

Name of Candidate: Artem Grebenko

PhD Program: Physics

Title of Thesis: CARBON NANOMATERIALS: SYNTHESIS AND CHARGE TRANSPORT MEASUREMENTS

Supervisor: Prof. Albert G. Nasibulin

The thesis document includes the following changes in answer to the external review process.

Prof. Maoshuai He:

1. In the table of contents, the first letter of all the words should be capitalized.

The table of contents was revised, first letters were capitalized

2. On page 13, “HipCO” is usually written as “HiPco” or “HiPCO”.

This typo was corrected

3. The writing of key words should be standardized, for example, in “single-walled carbon nanotubes”, a hyphen is needed between “single” and “walled”

The writing of key words was standardized.

4. On page 17, “arch-discharge” should be corrected as “arc-discharge”.

This typo was corrected

5. Spaces should be added where needed.

Space absence was double checked and corrected

6. On page 34, theta in XRD patterns should be written as “θ”.

This corrections was done

7. On page 43, “Figure 107” should be “Figure 10”.-1

This typo was corrected

8. On page 50, a full point is needed after 100 cm .

This typo was corrected

9. The format of the references should be carefully checked, and the volume number of some citation,
for example, reference 49, is missing.

I thank the reviewer for careful reading of the thesis. I have double checked all the references. All issues were corrected.

10. Please double check the title of the thesis, it seems there exists some inconsistency.

*Thesis title was modified to "C*ARBON NANOMATERIALS: SYNTHESIS AND CHARGE TRANSPORT MEASUREMENTS"

Prof. Dmitry Gorin:

1.Page 20, Figure 4, please add the scale bar;

Thank you for pointing out this issue. The figure is updated

2. Page 23, Figure 5, please add the description of Figure 5 d, e, f in the figure's caption;

Thank you for this correction. The caption was modified – each channel is associated with the corresponding text naming.

3. Page 24, Figure 6, please add the description of Figure6 a, b, c in the figure's caption;

Requested corrections are implemented.

4. Page 61, Figure 25, I didn't find any text related to Figure 25 B, C, D, E in the caption of Figure.
Please add scale bar for Figure 25 F, G, H; There is no Figure 25 K, as well as L, please remove
these symbols from Figure's caption;

Figure 25 was split into two new Figures 25 and 26 and corresponding captions were corrected.

5. Would be useful to apply backscattered electron scanning microscopy for characterization of
carbon nanotube surface doped by AuCl3

Thank you for this fruitful suggestion. However, we do not have these samples anymore and did not perform SEM backscattere electron analysis of them during investigation.

Prof. Vladimir Antonov:

P36 Last line in figure caption. " (I) Moire pattern…" What kind of this image, KPFM Image?

This is STM image obtained in ultra-high vacuum conditions.

P.37 Line 8 from the top. ” Most probably … The statement needs detailed explanation.

I appreciate careful reading of the manuscript and fruitful questions. However, we did not manage (me and collaborators) to find out the origin of single-crystal copper formation. We have a speculative hypothesis, that such alignment, which can be observed by all methods indicate in the thesis, originates from the high adsorption energy of CO molecules to the surface of transition metals.

P 42 Line 5 from the bottom. Would it possible to add arrows in the Fig 14d pointing at the SWCNT with changing potential

Arrows were added to the corresponding panels.

P43 Line 3 from the top. What kind of charge peculiarities author think of?

Text was modified to “These results were used to support spectroscopic findings concerning the charge transfer peculiarities, i.e. the observed additional absorption lines associated with tunelling processes and intertube crossings”

P47 Line 7 from the bottom. Statement “sequential tunnelling is forbidden…..” should be extendedwith
details.

This sentence is highly misguiding. It was changed to: “Sequential tunneling is forbidden inside the diamonds and current through the QD exists only due to high-order co-tunneling processes.”

P47 What is the motivation of the experiment of Section 4.1.4? Do you want to get some characteristics
of material?

Inititally, we intended to understand the possibility of balistic transport in the nanotubes produced by floating-catalyst technique. However, fabrication techniques that we have employed limited us due to high contamination of the fabricated devices. Therefore we finally focused on the investiagtion of various scenarios of electron tranposrt through a SWCNT QD in the regime of Coulomb Blockade

P48 Please decrypt colour coding of lines in Fig. 19.

The figure legend was largely improved. New version is:

“Figure 19. Shot Noise measurements in CB regime. Differential conductance plotted as a function of Vbg and bias voltage V. Lines indicate back gate voltage values and current ranges for which noise measurements were performed. Blue dots indicate current noise spectral density SI, differential conductance dI/dV is indicated by orange lines (corresponding axis are colored respectively). A,B and C indicate the cuts from Figure 18. Left columns are plotted versus source-drain voltage, while right panels are plotted versus I, and the region of plot is indicated by shaded region in the corresponding panel on the left (when applicable). Solid blue lines correspond to F =1, dashed lines to F = 0.5”

P48 The caption of Fig. 19. “…voltage ranges….” -> “…current ranges”

Corrected

P48 Line 2 from the bottom. What kind of resonance?

*Coulomb resonance. The text is modified to" The second, distinct scenario is observed when the Fermi level is shifted to the Coulomb resonance point (**panels b** and **b1**)."*

P49 Line 1 at the top. "…independent tunnelling processes". Explanation details are needed.

Explanation was expanded.

P49 Line 3 at the top "… these conditions..". He conditions should be clearly stated

Conditions were clarified.

P49 Last line "… different quantum states. .." . The states should be explained: are they dimensional
quantization, Coulomb blockade,…?

States were not specified. This is impossible due to a rather low quality of the particular device.

P50 Caption of Fig. 20 ".. (E-G)" to "…(F-H). Colour scale?

Not informative. There is no information in the ARPES lines intensity which is color coded. Only the position in terms of k and E makes sense.

Fig 20H: any details in the caption?

Indicated the reference in the caption.

P52 Line 7 from the top. Add commas :"…, like interference,.." . "..often occur.."

Corrected

P59 Line 9 from the top. " …of a 134 nm thick parafilm,.. "

It is correct, we used 134 micron thick parafilm layer. That is also supported by oscillations in Far infrared region of spectrum.

P55 line 2 from the bottom "…lateral…" →"…longitudinal…"?

Corrected.

P60 At which particular wavelength one would expect a benefit to have modulated reflection of SWCNT film?

Prominent phenomena can be observed in UV-vis-NIR ranges. The shorter wavelength is the higher is the reflection enhancement.

P62 Line 7 from the top. Would it be better to have somewhere a table of materials to

compare?

If I understand the question correctly, the table-wise comparison of differently synthesized graphene would not be highly informative due to the fact that typically graphene synthesis papers do not convey a deep and detailed chemical and structural analysis of the synthesized material.

P63 Line 3 from the top. “Microscopical analysis further helped to understand the contribution of the
defects and tunnel junctions into the FIR-THz ranges”. In which section these were discussed?

The utilization of KPFM to help interpreting the FIR-THz spectra are discussed in the beginning of the Chapter 4 and in details in papers 6,7 and 8 used in this thesis research.

Prof. Anton Andreev:

1. The super-poissonian noise in a single-carbon nanotube is an interesting phenomenon, can the author
comment a bit more on the nature of this observation? Is the author aware of shot-noise measurements
in graphene micro- and nanostructures? Can the super-poissonian noise be observed in graphene?
What is the typical value for the Fano factor of a ballistic graphene?

Super-poissonian noise is a pure intereference effect, that can not be observed, e.g., when the SWCNT is working in a Fabry-Perot regime. Due to the possibility of having a bundle-SWCNT sample and high contamination coming from the fabrication process we failed to give a deeper explanation of the observed large-F.

Graphene samples, both exfoliated and CVD synthesis were investigated my means of shot-noise measurements for, e.g. verification of the ballistic transport.

Yes, it can be observed, especially such situation is possible when a local maximum or minimum of conductance is considered.

0.3 is a tpyical value for the Fano Factor of a balistic graphene.

2. Can you please elaborate more on the distortion of the Coulomb diamonds?

We believe that the distortion of Coulomb diamonds is associated mainly with the fact that the investigated sample is not a single nanotube FET. It is supported by the following observations:

- *During fabrication, when the SWCNT was analyzed by means of AFM, we found out the height to be ~ 1.3 nm which is typically indicative of a bundle for the case of PeakForce QNM measurements.*
- *Individual SWCNT should have fluctuations of single-particle level spacing much smaller than the Coulomb energy defined by the capacitance of a quantum dot. It can also be indicative of a SWCNT bundle.*
- *However, the distortion can also be associated with the fact of operating at large negative Vbg values due to multi-subband transport.*

3. Can you please explain the discrepancy between quite low scattering rate and mobility values
extracted from the Hall effect measurements for the case of graphene FETs?

We associate this discrepancy with the procedure of graphene transfer. As far as during the preparation of Thz-spectroscopy compatible sample fewer harsh and destructive actions are taken with the sample, the observed scattering rates and associated mobilities are higher, than when compared to values extracted from FETs and Hall effect measurements.

Prof. Anvar Zakhidov:

1.What is the origin of a Lorentzian-like absorption line in THz graphene spectrum? Did the author conduct a research to understand its nature?

Indeed, together with the group of professor Boris Gorshunov we attempted to find the origin of this line, however, we have found it to be rather randomly appearing in experiment. It is not directly associated with defects, cracks and the size of crystallites. Most probably GHz spectroscopy should be performed alongside with THz and FIR regions to understand the nature of this absorption line. However, this research falls out of the scope of this particular dissertation. According to the latest published research such lines are visible from time to time and have not been clearly assigned to the sample state yet.

2. Can the suggested ultradensification patterning technique be transferred to SWCNT/MWNT forests?

Mos probably yes, it can be. However, author does not have an experience of AFM scanning of the SWCNT/MWNT forests. At the same time, I do not see any limitations, and even suppose, that densification also would result in altering of optical properties.

Prof. Nikolay Gippius

1. The abbreviation “AFML” in the title of Chapter 5 “AFML for SWCNT films” should be expanded to make it more clear for broad audience

Suggested correction was implemented.

2. The caption to Figure 25 (page 70) is a copy of that of Figure 24 and should be made adequate.

Figure was changed according to the request of another Jury member, and, as well, both new figures have an adequate caption.