Final report of grant OTKAPD 123927
Modeling of Molecular and Nanoscale Quantum Devices

Peter Rakyta

Introduction

During the three supported years of the grant we (i.e. the Principal Investigator (PI) of the grant with his supervised student and with his postgraduate colleagues) addressed several problems in physics of condensed materials and molecular systems. For example, (i) we investigated the quantum interference (QI) effects in three-terminal Andreev interferometers based on polyaromatic hydrocarbons (PAH's) under equilibrium and non-equilibrium conditions[1, 2], (ii) we constructed an interband tunneling picture to explain and predict the interlayer twist angle dependence of the induced spin-orbit coupling in heterostructures of graphene and monolayer transition metal dichalcogenides (TMDCs)[3] and investigated the conductance of edge states in gapped TMDC nano ribbons[4] , (iii) we studied the Cooper-pair splitting process induced by band structure engineering in graphene based multiterminal junctions[5], (iv) and examined transport processes through non-trivial magnetic structures in graphene-like ribbons[6]. Some of these problems were chosen to suit undergraduate skills, while other projects were started in the collaboration of international colleagues.

As a result of the grant the PI have successfully supervised three BsC students, his students participated in the Students National Scientific Conference and other scientific events organized for students and have won several prices.

In May of 2020 IBM announced the IBM Quantum challenge on the occasion of the fourth anniversary of the IBM Quantum Experience. During the experience the participants were given four problems to solve in four days. Although it was not part of the research plan, the PI of the present grant successfully participated in the challenge and earned an on-line badge [7] issued by IBM. Inspired by the 4th problem of the IBM Quantum challenge the PI started a new research project to develop a variational algorithm to decompose an arbitrary unitary operator into a quantum circuit of $U3$ and $CNOT$ gates. The PI with his colleagues developed a new algorithm, which can decompose any arbitrary unitary into a quantum circuit of fewer $CNOT$ gates than any other algorithm developed for the same purpose. For further details, see Sec. 3.

1 First year

During the first year of the supported period we studied the Josephson effect in gapped bilayer graphene based junctions. The gap opened by perpendicular electric field induces edge states in these systems, that are very robust against structural edge disorder. Our numerical simulations showed that the Josephson current in superconductor – bilayer graphene – superconductor junctions flows via these edge states, however tunneling bulk states can also contribute to the total Josephson current. These two contributions can be identified, for example, by the magnetic oscillations of the critical current. While the edge states show a SQUID-like oscillation pattern, the bulk tunneling states results in Fraunhoffer-like oscillations of the critical current. According to our reasoning, in realistic measurement one can observe an interplay of these two contributions. As reported in recent experiments the critical current indeed shows a SQUID-like oscillation pattern, with a higher oscillation peak at the center which (according to our reasoniung) originates from the tunneling states [8]. Our theoretical results may explain this experimental observation. The PI presented these result on the kick-off meeting of the FLAG-ERA JTC 2017 Topograph project of Engineering Topological Superconductivity in Graphene in order to strengthen the collaboration with experimental colleagues.

During the first year of the supported period we also started the studies of molecular junctions. When a single molecule is connected to external electrodes by linker moieties, the connectivity of the linkers to the molecular core can be controlled to atomic precision by appropriate chemical synthesis. Recently, the connectivity dependence of the electrical conductance and Seebeck coefficient of single molecules has been investigated both theoretically and experimentally. In our work we studied the connectivity dependence

of the Wigner delay time of single-molecule junctions. We also examined the connectivity dependence of superconducting proximity effects, which occur when the external electrodes are replaced by superconductors. Although absolute values of transport properties depend on complex and often uncontrolled details of the coupling between the molecule and electrodes, we demonstrated that ratios of transport properties can be predicted easily using tables of magic numbers, which capture the connectivity dependence of superconducting proximity effects and Wigner delay times within molecules. These numbers are calculated easily, without the need for large-scale computations. For normal-molecule-superconducting junctions, we find that the electrical conductance is proportional to the fourth power of their magic numbers, whereas for superconducting-molecule-superconducting junctions, the critical current is proportional to the square of their magic numbers. For more conventional normal-molecule-normal junctions, we demonstrate that delay time ratios can be obtained from products of magic number tables. We reported our results in Ref. [1].

Finally, we also started studies of three-terminal Josephson junctions. In this work we developed a theoretical model to obtain the Josephson current in a multiterminal setups accounting for both the bound and scattering states formed in the Josephson junctions. Using this model we examined the properties of multiterminal Josephson junctions and studied the possibility to turn them into a π-state regime by tuning the scattering properties of the junctions. These studied were motivated by schemes for the realization of quantum two-level systems (qubits) utilizing the phase shift of π between the superconducting banks in Josephson junctions. We also extended the numerical procedure to calculate the Josephson current under non-equilibrium conditions. We found that by inserting additional charge carriers into the system, the Josephson junction can be indeed driven into a π state. The BsC student of the PI presented the obtained results on the International Winterschool on Electronic Properties of Novel Materials.

2 Second year

In line with our research plan, during the second year of the supported period we implemented the diagrammatic non-equilibrium Green's function technique of Ref. [9] in the EQuUs software package. This technique can be further generalized to superconducting systems as well. With our code we reproduced the results of Ref. [10] regarding the transport process through a graphene quantum dot in the Coulomb blockade transport regime.

Also, we continued our studies of multiterminal molecular Josephson junctions. With the Keldysh non-equilibrium Green's function method we started to examine transport processes in such structures at finite bias voltage in the tunneling regime. In particular, we studied Andreev interferometers consisting of a PAH molecule coupled to two superconducting and one normal conducting terminals. We calculated the current measured in the normal lead as well as the current between the superconducting terminals under non-equilibrium conditions. We showed that both the QI arising in the PAH cores and the bias voltage applied to a normal contact have a fundamental effect on the charge distribution associated with the Andreev Bound States. QI can lead to an induced asymmetry in the spatial distribution of the electron- and hole-like quasiparticles. Also, the non-equilibrium charge occupation induced in the central PAH core can result in a π transition in the current-phase relation of the supercurrent for large enough applied bias voltage on the normal lead. The asymmetry in the spatial distribution of the electron- and hole-like quasiparticles might be used to split Cooper pairs and hence to produce entangled electrons in four terminal setups. We summarized our results in Ref. [2]. We also made the first steps to generalize our scope of interest to two-dimensional systems. We started to develop a code enabling us to investigate the transport properties of multiterminal two-dimensional normal-superconducting systems.

The PI of the grant together with his BsC student also started to study the transport properties of Transition Metal Dichalcogenide (TMDC) bilayers. These systems are interesting because of their large spin-orbit coupling and because of the valley degree of freedom. According to previous theoretical works, gapped bilayer graphene is expected to exhibit robust edge states (which was also confirmed by our previous results). We found that similar robust edge states are present in TMDC bilayers as well. We studied the band structure of these states and their connection to the Berry connectivity. (Since the studied system is time-reverse invariant, the total Berry curvature is zero.) We also studied the possibility to tune the properties of the edge states by an external electric field perpendicular to the sheet of the TMDC. We presented our result at several conferences in form of a posters or a short talks given by the student of the PI. The results were summarized in a BsC thesis written in Hungarian language [4].

In our other work we proposed an interband tunneling picture to explain and predict the interlayer twist

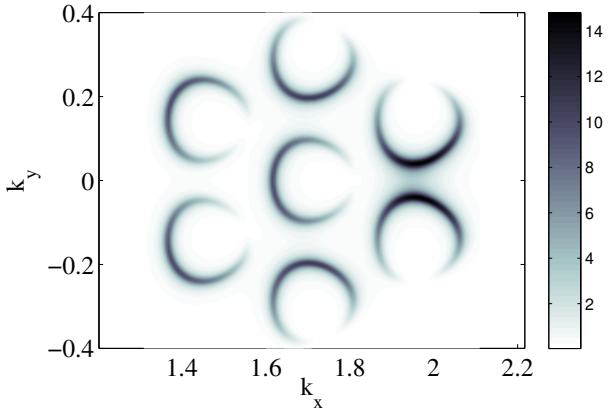

Figure 1: Theoretical calculation of the intensity distribution of the energy and angle resolved photoemission signal in the mini Brillouine zone for graphene on Ir(111) substrate close to the Dirac point. The six satellite Dirac cones around the central one have different orientations in line with the preliminary experimental results. The units of the k_x and k_y wave vector components are given in units of the lattice constant, and the photoemission signal is given in arbitrary units in the colorbar indicating only the high and low intensity directions.

angle dependence of the induced spin-orbit coupling in heterostructures of graphene and monolayer TMDCs. We obtained a compact analytic formula for the induced valley Zeeman and Rashba spin-orbit coupling in terms of the TMDC band structure parameters and interlayer tunneling matrix elements. We parametrize the tunneling matrix elements with few parameters, which in our formalism are independent of the twist angle between the layers. We estimate the value of the tunneling parameters from existing DFT calculations at zero twist angle and we use them to predict the induced spin-orbit coupling at non-zero angles. Provided that the energy of the Dirac point of graphene is close to the TMDC conduction band, we expect a sharp increase of the induced spin-orbit coupling around a twist angle of 18 degrees. We reported our results in Ref. [3].

We also started a collaboration with experimental colleagues at Helmholtz Zentrum in Berlin. We were given an opportunity to participate in a project examining the angle resolved photoemission signal measured on a graphene-Ir(111) heterostructure. This system exhibit a nontrivial electronic structure due to the moire interference pattern between the graphene and Ir lattices. In particular, around a central (gapped) Dirac cone another six satellite Dirac cones appear at the high symmetry point of the mini Brillouine zone. Based on our experiences gained in our previous work [11], we developed an effective model to describe the band structure of the graphene-Ir(111) system and we also calculated the intensity distribution of the angle resolved photoemission signal (see Fig.1 to see our results). Our theoretical results obtained so far were in decent agreement with the experiments. Unfortunately, we were informed about a faulty experimental equipment by our colleagues working at the Helmholtz Zentrum and the project remained stalled since then.

3 Third year

During the third year of the supported period a fourth BsC student joined our team and started to study the transport processes in the Coulomb blockade transport regime. The objective of the research is to examine the coupling between the Hall edge states (induced by applying perpendicular magnetic field) and a quantum dot located in the center of the scattering region. We want to examine the signatures of the central quantum dot in the transport properties measured on the Hall edge states. Our work is motivated by experimental results of Ref. [12]. In our theoretical model, however, we are using two dimensional electron gas (2DEG) as a central region. We decided to use 2DEG in our calculations since in 2DEG it is much easier to create quantum dots (at least in our numerical simualtions) via electrostatic effects. So far we have implemented all the physical effects in our theoretical model, and just started to run the numerical simulations on the studied system.

We also started studies of graphene based Cooper pair splitting devices based on different geometrical considerations. Motivated by the band structure engineering approach described in Ref. [13] we created differently doped regions in the graphene junction connected to superconducting banks. In contrast of the

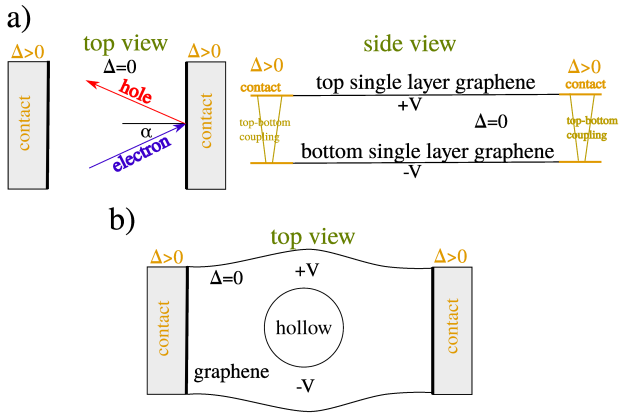

Figure 2: The two geometries used in our calculations to model the Cooper pair splitting process. The spatial splitting of the electron pairs is assisted by different electrostatic properties in regions indicated by the $+V$ and $-V$ doped areas.

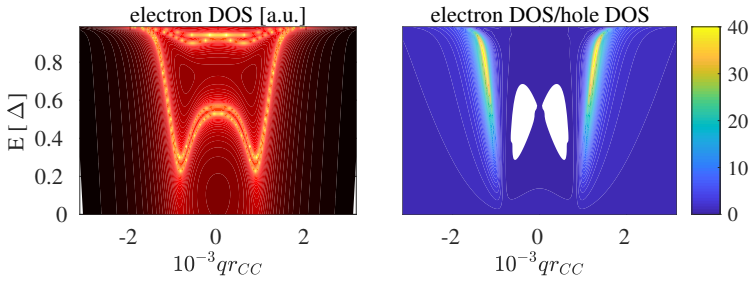

Figure 3: Left: The density of states corresponding to the Andreev bound states in a geometry, where two single layer graphene layers are placed on top of each other. Right: the ratio of the density of states calculated for the electron and hole-like parts of the wave function in the upper leyer. Bright areas indicate regions where the electron-like component are dominant. (In the bottom layer these areas correspond to hole-dominant regions.)

basic geometry described in [13], we examined systems including two superconducting contacts instead of one. According to our expectations, in these systems the (resonant) Andreev bound states can host spatially separated (but still entangled) electrons correlated by the crossed Andreev reflection happeninng on the surface of the superconducting banks. We expect that connecting normal contacts to regions where the Cooper pairs become spatially separated, the signal coming from the crossed Andreev reflection might be significantly increased. (This expectation is based on our previous results obtained on molecular junctions [2].) We proposed to examine this effect on two distinct geometrical designs and started numerical simulations with two MsC students who have previously passed their BsC exams. The two designs are briefly described in Fig. 2. In the first case two single layer graphene ribbons are aligned on top of each other and connected to two superconducting contacts. The top layer is tuned to electron doping and the bottom layer is tuned to hole doping. In the second case we formed a two-armed interferometer from a graphene layer connected to two superconducting banks. In this case the two arms of the interferometer are tuned to host different (i.e. electron- or hole-like) charge carriers. Similarly to the reasoning of [13], in these systems we also expect the spatial separation of the electron and hole-like parts of the Andreev bound states. Although these studies are still in progress, we already have reached some preliminary results. Some of these results are plotted in Fig. 3. In this figure we plotted the spectral density of the Andreev bound states and the ratio of the electron-like and hole-like components of the spectral density in the upper layer of system depicted in Fig.2.a). We can see, that for certain transverse momentum values q the electron density is much higher than the density of the hole-like quasiparticles. Similarly, in the bottom layer the hole-like components will be dominant at the same parameters regions. These results indicate the spatial separation of the electron- and hole-like quasiparticles for specific parameters. In the next step we aim to connect two additional normal contacts to the system at points where the spatial separation occurs and we calculate the non-local differential conductance to see the efficiency of the Cooper pair splitting process.

Finally, as was mentioned in the introduction, the PI of the grant has launched a new project motivated

by the fourth problem of the IBM Quantum Challenge 2020. We developed a new heuristic algorithm to decompose an arbitrary unitary into a sequence of $U3$ single qubit and $CNOT$ two qubit operations. As stated by theoretical studies there is a general lower bound on the number of $CNOT$ operations needed to decompose an arbitrary unitary. This lower bound scales with $N_{CNOT} \sim \frac{1}{4}4^n$, where n is the number of qubits described by the unitary operator. However, currently there is no working algorithm reaching this theoretical lower bound. Instead, the best known algorithm (called Quantum Shannon Decomposition) implemented in quantum computer services today is scaling with $N_{CNOT} \sim \frac{1.9}{4}4^n$[14]. Our approach surpasses this algorithm, the number of $CNOT$ operations needed for the decomposition of an arbitrary unitary scales as $N_{CNOT} \sim \frac{1.25}{4}4^n$ in our algorithm. While, we currently preparing a manuscript to summarize our results, the implementation of the algorithm can be accessed only via a private GitHub repository.The implementation is also supported by web-based source code documentation that can be temporary accessed via Ref. [15]. Although it was not part of the original proposal, the PI really enjoyed participating in this project.

4 summary

In summary, during the supported period three students completed their BsC program under the supervision of the PI of the grant. The scientific results of the grant have been reported in three peer-reviewed journals and were presented in forms of conference talks and posters by the PI himself and by his supervised students. We currently preparing a manuscript to report our results regarding the gate decomposition of arbitrary unitaries. The algorithm developed by the PI of the grant surpasses all currently available algorithms, thus might by of high importance in the field of quantum information technology. Also, we continue our work on studying the Cooper splitting processes in graphene or 2DEG based junctions. Currently we are working on three concurrent projects with promising preliminary results outlined in Sec. 3.

References

[1] P. Rakyta, A. Alanazy, A. Kormányos, Z. Tajkov, G. Kukucska, J. Koltai, S. Sangtarash, H. Sadeghi, J. Cserti, and C. J. Lambert, J. Phys. Chem. C, **123(11)**, pp 6812-6822 (2019).

[2] N.L. Plaszkó, P. Rakyta, J. Cserti, A. Kormányos, and C. J. Lambert, Nanomaterials **10(6)**, 1033 (2020).

[3] A. David, P. Rakyta, A. Kormnyos, G. Burkard, Phys. Rev. B **100**, 085412 (2019).

[4] E. Zsurka: *Élállapootk kétrétegű átmenetifém-dikalkogenidekben*, BsC thesis (2019).

[5] P. Rakyta: *Crossed Andreev reflection in layered graphene structures*, Topograph meeting (2020).

[6] B. Farkas: *Grafén vezetőképessége nemtriviális topológiájú mágneses terekben*, BsC thesis (2020).

[7] https://www.youracclaim.com/badges/b5d8b5e8-2d74-4d03-a12a-3bbfbac019a0/embedded

[8] M. J. Zhu, A. V. Kretinin, M. D. Thompson, D. A. Bandurin, S. Hu, G. L. Yu, J. Birkbeck, A. Mishchenko, I. J. Vera-Marun, K. Watanabe, T. Taniguchi, M. Polini, J. R. Prance, K. S. Novoselov, A. K. Geim and M. Ben Shalom, Nature Communications **8**, 14552 (2017).

[9] H. Scholler and G. Schön, Phys. Rev. B **50**, 24 (1994).

[10] I. Weymann, J. Barnaś, and S. Krompiewski, Phys. Rev. B **92**, 045427 (2015).

[11] P. Rakyta, A. Kormányos, J. Cserti, Phys. Rev. B **83**, 155439 (2011).

[12] E. Tóvári, P. Makk, P. Rickhaus, C Schönenbergerb and Sz. Csonka, Nanoscale **8**, 11480 (2016).

[13] J. Cayssol, Physical Review Letters, **100**, 147001 (2008).

[14] R. Iten, R. Colbeck, I. Kukuljan, J. Home, and M. Christandl, Phys. Rev. A 93, 032318 (2016).

[15] https://itl88.elte.hu/rakytap/Download/QGD/html/