Engineering topological superconductivity in graphene

OTKA-NN 127903

Topograph FlagERA project

Összefoglaló

A TopoGraph projekt célja az volt, hogy lépéseket tegyen a topologikus szupravezetés megvalósítása felé grafén alapú van der Waals heterostruktúrákban. A Topograph projektben kifejlesztettük és megvizsgáltuk a Majorana zéró modusokat potenciálisan magában foglaló heterostruktúrák különböző összetevőit: Alagútszondákat terveztünk és alagútspektroszkópiát mértünk nem egyensúlyi körülmények között. Kifejlesztettünk egy kétrétegű grafén rendszert is, ahol szupravezető áram-fázis összefüggést mértünk, és kvantum spin-Hall állapotot figyeltünk meg nagy mágneses tereknél. Ez a rendszer az egyik legkisebb magnetométer, amelyet eddig létrehoztak.

A projekt egyik legnagyobb feladata az volt, hogy kísérletileg és elméletileg feltárja a spin pálya kölcsönhatások szerepét és típusát a TMDC/grafén heterostruktúrákban, különböző elrendezésekben. Sikeresen megmutattuk a spin pálya csatolás hidrosztatikus nyomással történő növelését, és kiszámítottuk a rétegek közötti csavarodási szögtől való függését. Megmértük az áram-fázis relációt grafénben indukált spin pálya mellett. A kétrétegű grafénben, amelyet a WSe2 kristályok közé szendvicseltünk, anomális fázist figyeltünk meg, és a szuperáram méréseknél a topologikus természetének jelei láthatók.

A projekt eredményeként 2 BSc és 4 MSc szakdolgozat, valamint 4 TDK dolgozat született. Az eredmények 20 kutatási cikk publikálását (vagy elfogadását) eredményezték vezető folyóiratokban, köztük egy Phys. Rev. Lett és több Nano Letters.

Hasznosulás/Utilization

Bár az általunk folytatott kutatás alapkutatás jellegű, és nem közvetlen ipari kapcsolatokon keresztül hasznosulnak, a grafén alapú elektronikai eszközökben komoly potenciál rejlik. Ezt jól mutatja a Graphene Flagship, az EU zászlóshajó projektje, ami a kutatási eredmények ipari alkalmazását tartja egyik fő céljának. A projekt egyik kiemelkedő eredménye ebben az értelemben a projekt keretében kifejlesztett kétrétegű SQUID, amely az egyik legkisebb létező magnetométer.

Though our project is a fundamental research project without direct interaction with industrial partners, there is a huge potential in graphene based electronic circuits. This is well demonstrated by the Graphene Flagship projects, where one of the main goals is to find industrial applications to graphene based electronic circuits. One of the highlights of the project in this sense is the double layer SQUID developed partially within this project, which is one of the smallest magnetometers out there. This was also highlighted in several media appearances.

Summary

The FlagERA project targeted the exploration of topological states (Majorana Zero Modes, MZM) in graphene based structures coupled to superconducting electrodes. This project leverages the unique properties of graphene: long-range ballistic transport, clean interfaces with superconductors, tuneable properties through van der Waals engineering. The aim of the TopoGraph project was to make steps towards engineering topological superconductivity and MZMs in graphene-based van der Waals heterostructures.

In the Topograph project we developed and investigated the different ingredients for heterostructures hosting MZMs: Tunnel probes were engineered and tunnel spectroscopy was performed under non-equilibrium conditions. We also have developed a double-layer graphene system, where we measured superconducting current-phase relation and observed a quantum spin-Hall state at large magnetic fields. This system is one of the smallest magnetometers that exists up to now.

The largest effort in the project was to reveal the role and type of spin orbit interaction in TMDC/Graphene heterostructures in different layouts both experimentally and theoretically. We have demonstrated the boosting of spin orbit coupling with pressure and calculated its dependence on the twist angle between the layers. We have measured the current phase-relation in graphene with induced spin orbit coupling. In bilayer graphene, that was encapsulated between WSe2 crystals, an inverted phase was observed and in supercurrent measurements signatures of its topological nature was seen.

The project resulted in 2 BSc and 4 MSc thesis and 4 two theses for the Hungarian Scientific Students' Associations competition (TDK). The results have resulted in the publication (or acceptance) of 20 research papers in leading journals, including a Phys. Rev. Lett, and several Nano Letters papers.

The project resulted in new research collaborations, that are ongoing after the project also: with the group of Srijit Goswami in TU Delft, and Pablo San-Jose in Madrid (CSIC).

Below, we give a short overview of the main results of the project, concentrating on the results of the BME node.

Spin orbit studies

Tuning spin orbit with pressure cell. We have found before the project that in a single layer graphene placed on WSe_2 a large induced spin-orbit interaction appears, which contains a Rashba and a valley-Zeeman type interaction. However, it also strongly depends on the distance between the two sheets, since the proximity effect relies on the overlap of the orbitals of the two materials. Therefore, by tuning the interlayer distance it is expected, that the strength of the SOC can change. In our pioneering work the we have engineered a setup, where a nanodevice bonded on a circuit board can be placed inside a pressure cell and using hydrostatic pressure the 2D layers of the nanodevice can be pressed closer to each other. Therefore, we have started to investigate how the spin orbit interaction depends on the distance between the layers. In our pioneering work we have engineered a setup, where a nanodevice bonded on a circuit-board can be placed inside a pressure cell and using hydrostatic pressure the 2D layers of the nanodevice can be pressed closer to each other. We have tested this on a graphene device on a TMDC substrate, where at 1.5K no sign of weak anti-localization and hence of SOC have been seen. As the pressure was gradually increased (in several cool-downs) a weak anti-localization peak appeared and grown stronger with every step. The results are shown in Figure 1. and was published in B. Fülöp et al., npj 2D Materials and Applications 5, 82 (2021) and in B. Fülöp et al., Journal of Applied Physics 130, 064303 (2021).

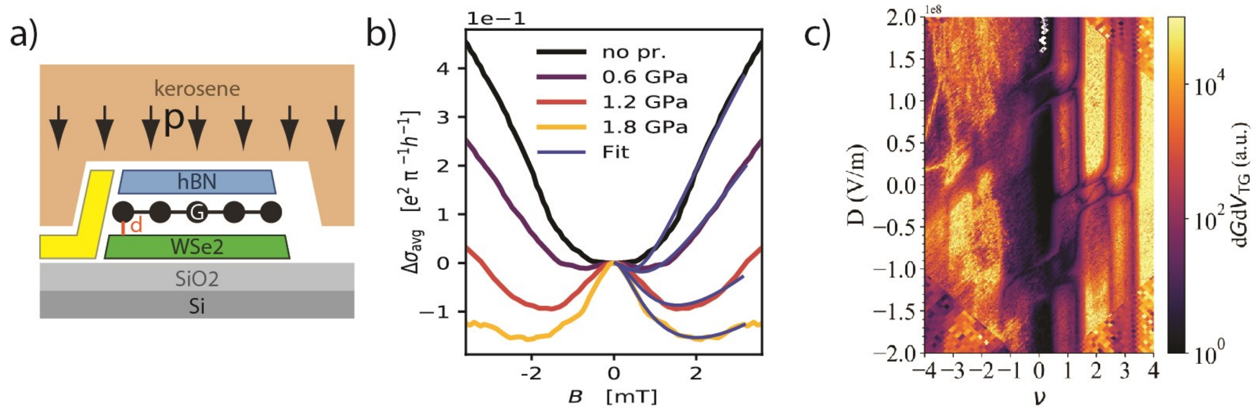

Figure 1: a) Sketch of graphene/SOC structures. b) Weak anti-localization measurement on a graphene/WSe2 structures as a function of hydrostatic pressure. At zero pressure no sign of spin orbit interaction is present, which appears at larger pressures. c) Differential resistance of a BLG/WSe_2 structure as a function of displacement field and filling factor at B=14T. The Landau level crossings give information on the strength of the SOC coupling.

Bilayer graphene (BLG) and proximity spin-orbit. We have also investigated *bilayer graphene in proximity to WSe_2*. The graphene was placed on a single layer of WSe_2, and this structure was encapsulated between hBN flakes. To increase the quality of the devices we have placed the stack on a graphite gate. We have observed *weak anti-localization* which signals the presence of SOC in graphene. We also investigated the strength of SOC using QHE effect measurements. It was recently shown that by investigating *Landau level crossings* as a function of electrical fields the strength of SOC can be obtained (D. Wang et al., Nano Lett., 19, 7028, 2019). Our measurements (BME node) revealed a valley Zeemann (Ising) spin orbit coupling ~1.2 meV. We have also observed an increase in the SOC using pressure for BLG as well (1.7 meV). This study is still ongoing.

Band inversion from SOC.

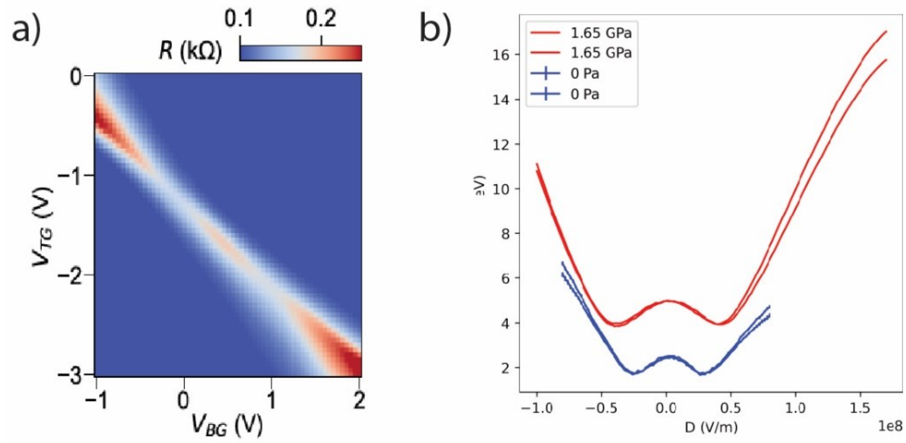

Figure 2: a): Differential resistance as a function of top and bottom gate voltages in a WSe2/BLG/WSe2 heterostructure. The bright colours show gaps at the charge neutrality point. At large voltages the displacement field opens up a gap, whereas at low displacement fields the SOC opens a gap. b) Extracted gaps as function of displacement field for ambient and high pressure. The curves show a clear increase of the gaps around zero displacement field.

If BLG is encapsulated between two WSe_2 flakes, that are rotated close to 180 degrees, a special band structure forms. In this case the Valley-Zeemann coupling is opposite in the two layers, which effectively will realize the Kane-Mele Hamiltonian and the system becomes gapped at the charge neutrality point. This gap can be closed and reopened with electric field. We have observed this phase in several samples (together with the TU Delft node), and in one we have managed to perform pressure experiments in order to boost the strength of SOC (see Fig. 2.). We have observed a more robust inverted phase, which we have also verified using thermal activation measurements. Finally, at large magnetic fields signatures of the SOC was visible in the LL spectrum. This work is being written up.

Theory of the angle dependence of SOC. It has been known, that if graphene is placed on TMDC substrates SOC of the TMDC is inherited in the graphene. However, it was an open question if the strength of different spin orbit terms depends on the alignment angle between graphene and the TMDC lattice. In collaboration with the group of G. Burkhard in Konstanz we have investigated this for the case of MoS2. They have developed a methodology which works in the momentum space and takes into account how the bands of the TMDC and the orbitals of graphene are hybridized. The beauty of the method is that a semi-analytical form has been achieved for the spin orbit parameters as a function of the twist angle. This means that from the knowledge of the spin-orbit parameters for one particular twist angle, the parameters for the rest of the angles can be obtained. This single angle can be calculated with DFT supercell method. It can be clearly seen that both the Rashba and the valley Zeeman spin orbit terms depend on the rotation angle, and magic angles where these terms are maximized can be found. These results could also explain the differences between different experimental studies in the field, which were likely all conducted at different rotation angles.
Furthermore, we have investigated the case of single layer graphene encapsulated by TMDCs from both sides. We have shown that the Rashba SOC can be affected by quantum interference from the TMDCs on the bottom and top. This interference affects both the spin polarization in graphene, both charge to spin conversion effects. These results have been published in A. David et al., Phys. Rev. B 100, 085412 (2019) and Cs. G. Péterfalvi et al., arXiv:2111.02781 (accepted for publication in Physical Review Research).

CPR measurements

One of the main goals of the project was to study current phase relation (CPR) in graphene with induced SOC. This was done by embedding the Josephson junctions in an asymmetric SQUID, where the modulation of the switching current originates from the CPR of the Josephson junction with smaller critical current. We have studied different structures including WSe2/SLG/WSe2, WSe2/BLG/hBN, WSe2/BLG/WSe2, hBN /SLG/hBN.

These studies included a lot of technical development, including proper correction for inductance etc. artifacts. Out of these structures for WSe2/BLG/WSe2 we have found an anomalous current phase relation. When the junction was placed in in-plane magnetic fields a large phase shift appeared, on the order of 2π (!), hence forming a φ_0 junction (Fig. 3.). The phase is gate tuneable, which allows us to do a proper phase comparison. This is one of the main results of the project, however the origin of this effect unknow – our initial estimates would predict phase shifts that are an order of magnitude smaller. We are still doing some follow-up experiments before publishing these results.

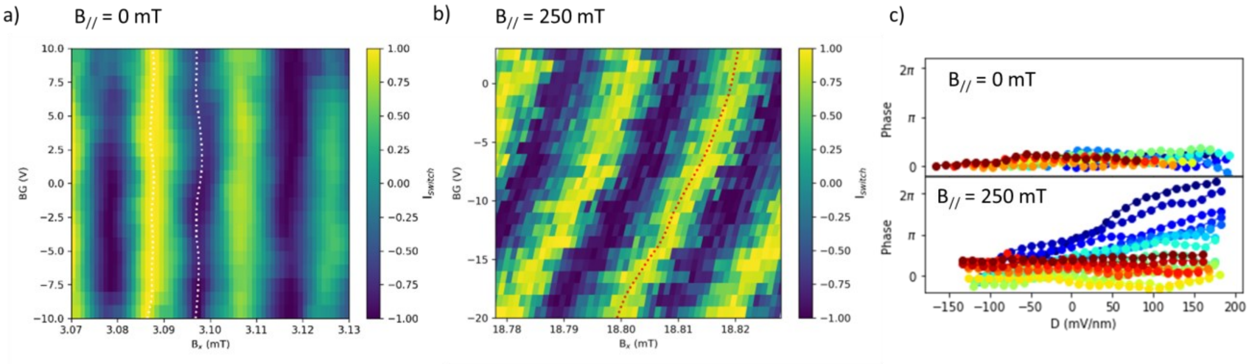

Figure 3: Large φ_0 phase for WSe2/BLG/Wse2. Switching current (modulation) as a function of back-gate and external magnetic field for 0T (a) and for 250mT (b). c) Extracted phases as a function of displacement field for different densities (colours). A large gate tuneable phase is visible in finite magnetic fields.

Double layer graphene

Together with the university of Basel - we have worked on Josephson junction that are made of two parallel graphene sheets, that are separated by hBN (the structure: Graphite gate/hBN/SLG/hBN/SLG/hBN/topgate). We have verified that supercurrent flows in both layers, and we could perform CPR measurements, where the control parameter was the in-plane magnetic field between the layers (Fig. 4.). This method has the advantage that no orbital effects or interference is present in the measurements. We have found moderately skewed CPR, with larger transparency at high doping. In high magnetic fields we have formed Landau levels, and observed splitting of the lowest Landau level. By setting ± 1 filling factors in the top and bottom graphene sheets we have managed to realize an artificial quantum spin-Hall state. This part of the work has already been published. We did not observe however supercurrent in this regime yet. We are still trying to optimize both the structure and the setup to have enough supercurrent in the topological regime. The CPR measurements have been published in D. Indolese et al., Nano Lett., 20, 7129 (2020).

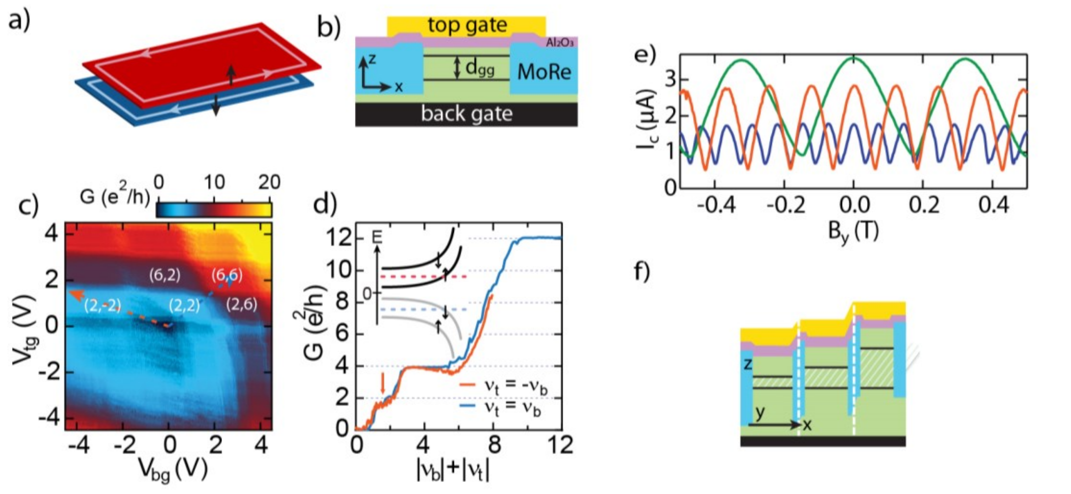

Figure 4: Measurements on the double layer setup. a) Sketch of artificial QSHE. b) Device sketch. c) Conductance as a function of the top and bottom gate voltage at 5T. d) Cuts from c) shown by the dashed

line. The arrow shows the QSHE (see inset for band structure). e) Critical current as a function of in-plane magnetic field. The different colours correspond to different hBN spacers as shown in panel f).

Tunnel spectroscopy

We have developed tunnelling spectroscopy into graphene. We have used single and bilayer CVD hBN barriers and Pb superconducting electrodes as spectrometers. The samples were prepared on stripes of graphene, where 3 terminals were fabricated. The outer ones were normal leads, whereas the middle one was the spectrometer. We have fabricated samples with different sizes (~1-100um length). We have measured the tunnelling spectrum (conductance vs. bias voltage) for different gate voltages. We have also recorded such curves, when a heating bias voltage was applied between the two normal contacts, which drives the graphene out of equilibrium. Using the tunnel measurement, we have extracted the non-equilibrium distribution of graphene for different gate voltages and heating biases. A thorough investigation of the extracted distributions functions was done and from this different electron cooling mechanisms were observed. For the smaller samples electron-electron interaction and escape through the contact is the dominant cooling mechanism for longer samples electron-phonon coupling, and cooling via the phonons become more important. We have also seen signature of a double-step distribution function. There results were published in PRB (S. Zihlmann et al., PRB 2019).

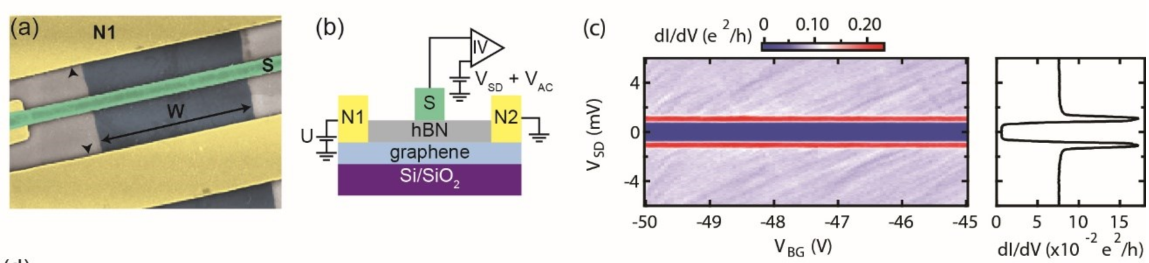

Figure 5: Tunnel spectroscopy measurements. a) Device outline. Blue is graphene, green is the SC tunnel contact, whereas yellow contacts are made from normal metal. b) Measurement scheme. c) Differential conductance (without heating bias) as a function of bias and gate voltage. By performing and averaging over the gate voltage to reduce UCF, the DOS of the SC is recovered.

Superlattice studies

We have managed to fabricate clean graphene Josephson with a superlattice. The superlattice was realized by aligning the graphene lattice to the hBN lattice, thus realizing a moiré structure. We have found that the supercurrent is decreased around the secondary Dirac points formed by the moiré superlattice. Moreover, taking advantage of the diffusive nature of the sample, and using both the supercurrent and normal state measurements we could extract the density of states in the sample. Moreover, using magnetic field measurements the current distribution in the sample could be extracted. We have found a substantial edge current contribution which we attributed the doping of the graphene edge (likely originating from plasma etching). These results were published in Phys. Rev. Lett. (D. Indolese et al, PRL 2018). We have also realized super-superlattice (without superconducting contacts), where both the top and bottom layer of the hBN was aligned with graphene resulting in the superlattice of the two underlying moiré (L. Wang et al., Nano Lett. 2019).

Finally, hydrostatic pressures in the range of 2GPa can substantially change the band structure of twisted graphene structures. We demonstrated this on twisted doubly bilayer graphene, two bilayer graphene layers place stacked together with a rotation angle of 1.07, close to the magic angle. At this small rotation angles, the two layers strongly hybridized and flat bands, that are electric field tunable, arise at low energies. These narrow bands are separated from higher lying bands by moiré gaps. Our experiments have shown that a pressure of 2GPa is enough to close these bandgaps in the spectrum, and substantially altering the spectrum of the system (Fig. 6). This was in good agreement with our calculations. The work was published in Nano Letters (B. Fülöp et al., Nano Lett., 21, 8777 (2021)).

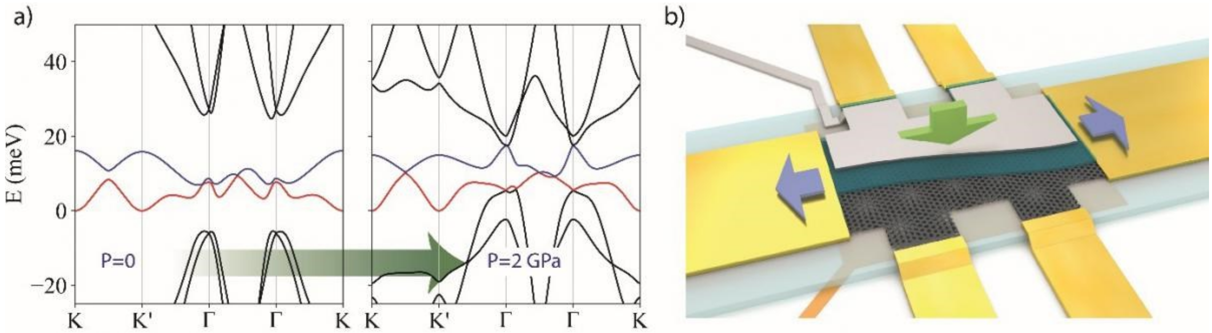

Figure 6: a) Band structure of twisted double bilayer graphene (with twist angle of 1.07) at ambient and large pressures. A strong modification of the bandstructure is observed. b) Artistic view of a device, with the green arrow showing the hydrostatic pressure.

Major investments

During a project a larger investment was the purchase of a low temperature sample holder that can be inserted to the dilution fridge to 20 mK. This probe allows fast cooldown of samples from room temperature to 20mK in less than a day. This is used for CPR measurements, along with the small electronics components that were purchased. Smaller developments included the development of a new 4K sample holder for the pressure cell setup and smaller upgrade of the micromanipulator assembly for heterostructures fabrication.

Parallel research

The running costs of this projects heavily exceeds the budget of the current proposal. The PI has received his salary from an OTKA PD project, then later from a Marie Curie and the Bolyai grant. Some of the research was funded by the Nanoelectronics momentum grant of Szabolcs Csonka and some of the measurements were funded by the partners from abroad, where the measurements were done. Some of the work was started before the project, when the PI was a postdoc in Basel. In parallel to this project the PI was the coordinator of an OTKA FK grant (123894), which focused on electron optics in graphene, spin injection, strain studies etc. [2, 6-9]. Very recently the PI was awarded the momentum grant which focuses on the strain and pressure studies of moiré superlattices. Andor Kormányos also started to coordinate an OTKA-K grant (134437), where the focus in on theoretical studies and where the theory part of the work in Ref. 14 was done, and also Ref. 16 is a part of that project.

We note that this project has been extended one year longer due to COVID period (without additional funding) in order to facilitate collaboration within the FlagERA consortium.

Publications (connected to the project)

Thesis

- Szentpéteri Bálint, TDK thesis - *Nem-lokális spintranszport mérések grafén/BiTeBr heteroszerkezetekben*
- Kocsis Matyas, TDK thesis- *Tuning the Rashba parameter in BiTeBr with ionic-liquid gating*
- Kedves Máté, TDK thesis - *Towards engineering topological states in graphene*
- Márffy Albin, BSc thesis- *Van der Waals heteroszerkezetek vizsgálata nyomás alatt*
- Márffy Albin, TDK thesis - *Kétdimenziós nanoáramkörök vizsgálata nyomás alatt*
- Szentpéteri Bálint, MSc thesis - *Spin fizika kétdimenziós heteroszerkezetekben*
- Kedves Máté, MSc thesis – Quantum effects in graphene
- Márffy Albin, MSc thesis - *Van der Waals heteroszerkezetek létrehozása és vizsgálata nyomás alatt*
- Pápai Tamás, BSc thesis - *Szupravezető áram-fázis reláció grafén heterostruktúrában*
- Pápai Tamás, MSc Thesis - *Investigation of Graphene Josephson Junctions*

Research papers

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   https://aip.scitation.org/doi/10.1063/5.0058583
4. Bálint Fülöp, Albin Márffy, Simon Zihlmann, Martin Gmitra, Endre Tóvári, Bálint Szentpéteri, Máté Kedves, Kenji Watanabe, Takashi Taniguchi, Jaroslav Fabian, Christian Schönenberger, Péter Makk, Szabolcs Csonka, *Boosting proximity spin orbit coupling in graphene/WSe2 heterostructures via hydrostatic pressure*,
   https://www.nature.com/articles/s41699-021-00262-9
5. Wang, Lujun ; Zihlmann, Simon ; Baumgartner, Andreas ; Overbeck, Jan ; Watanabe, Kenji ; Taniguchi, Takashi ; Makk, Péter ; Schönenberger, Christian, *In Situ Strain Tuning in hBN-Encapsulated Graphene Electronic Devices,* Nano Letters 19, 4097 (2019), IF:12.3
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6. Wang, Lujun ; Makk, Péter ; Zihlmann, Simon ; Baumgartner, Andreas ; Indolese, David I. ; Watanabe, Kenji ; Taniguchi, Takashi ; Schönenberger, Christian, *Mobility Enhancement in Graphene by in situ Reduction of Random Strain Fluctuations*, Phys. Rev. Letters., 124, 157701 (2020), IF:8.4

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14. Alessandro David, Péter Rakyta, Andor Kormányos, and Guido Burkard: *Induced spin-orbit coupling in twisted graphene–transition metal dichalcogenide heterobilayers: Twistronics meets spintronics*, Phys. Rev. B. 100, 085412 (2019) https://journals.aps.org/prb/abstract/10.1103/PhysRevB.100.085412
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