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## Report on the dissertation

## Synthesis and electron spin structure of quasi-two-dimensional systems with a combination of spin-orbit and magnetic exchange interactions

## by Artem G. Rybkin (St. Petersburg State University)

The dissertation explores how the spin-orbit properties of graphene can be controlled via its interaction with various substrates, particularly heavy metals like gold and platinum. These materials induce strong spin-orbit coupling in graphene, enabling access to novel spin structures and topological effects.

The amount of experimental work is impressive. A wide range of graphene systems was synthesized using multiple techniques and characterized by advanced spectroscopy and microscopy, including STM, ARPES (also spin- and time-resolved), LEED, and SQUID, both in-house and at synchrotron facilities. The integration of synthesis with in-depth spectroscopic analysis is a major strength.

The results significantly advance the development of spintronic materials and devices. Three device concepts are proposed—a graphene spin filter, a SOT-MRAM cell, and a mid-infrared detector of circularly polarized radiation—supported by three filed patents.

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### Chapter 1 – Current state of research

This chapter reviews the theoretical foundations and recent progress in controlling spin structures in quasi-two-dimensional systems. It highlights key mechanisms such as spin-orbit interaction, the magnetic proximity effect, and topological electronic phases, with particular emphasis on graphene and topological insulators as model materials.

The chapter is comprehensive, cohesive, and well written. Since spin-related transport effects such as QAHE are invoked throughout the discussion (e.g., in connection with Rashba splitting and proximity effects), the author might consider opening the chapter with a review of such transport phenomena. It could also be valuable to mention the recent work in *Phys. Rev. X* **10**, 041013 (2020) on transient topological states in graphene observed via circular-dichroism ARPES. Additionally, while the approximation of ARPES final states as plane waves is widely used, the author may wish to note that non-free-electron effects can arise not only at low excitation energies but may also persist at high photon energies well above 1 keV (*Nature Comm.* **14**, 4827 (2023)).

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**Chapter 2 – Methods**

The author describes the experimental and computational approaches used throughout the work, including spin- and angle-resolved photoemission spectroscopy (ARPES), X-ray photoelectron spectroscopy (XPS), scanning tunnelling microscopy (STM), and density functional theory (DFT). Detailed procedures for sample preparation and surface cleaning are also presented. The chapter is comprehensive and to the point.

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**Chapter 3 – Induced spin-orbit interaction in thin metal layers**

The chapter investigates thin Al layers deposited on W(110) substrates and shows that quantum well states in the Al layer exhibit spin splitting due to hybridization with substrate states. The results demonstrate that strong spin-orbit coupling from the substrate can be effectively transferred into non-magnetic overlayers.

The chapter is clearly structured and well written. In Fig. 3.2(b), the author might consider indicating the bulk bands originating from the substrate. It would also be helpful to specify the photon energy used and the corresponding out-of-plane momentum. Furthermore, the hole-like feature marked as QWS1 seems unusual for a quantum-well state and may warrant further justification. In Fig. 3.7(e,g), it would be interesting to clarify the nature of the weaker QWS-like dispersions seen below QWS1.

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**Chapter 4 – Magnetically doped systems with strong spin-orbit interaction**

Magnetically doped topological insulators are studied using both static and pump-probe photoemission techniques. The author shows that magnetic doping modifies the electronic structure and leads to a pronounced two-dimensional photovoltaic effect that depends on the Dirac point position.

The chapter is consistent and interesting. It may be a good place to mention soft-X-ray resonant ARPES and muon spin spectroscopy studies of V-doped $Bi_2(Se,Te)_3$, which revealed a large impurity density of states near the Dirac point that impairs the QAHE. Related phase separation effects observed in those studies could also be briefly considered. Also, the author discusses the “phonon bottleneck” as a mechanism explaining slow relaxation of TSSs in V-doped $Bi_2(Se,Te)_3$. As I understand it, this mechanism relates the slow relaxation to a small Luttinger area near the Dirac point, which limits the available phase space for phonon scattering of Fermi electrons. However, in Fig. 4.2, the slowest relaxation is observed for $Bi_{1\cdot97}V_{0\cdot03}Te_{2\cdot4}Se_{0\cdot6}$, which appears to have the largest Luttinger area—including contributions from bulk conduction-band states. Are there other mechanisms that could explain this inconsistency?

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**Chapter 5 – Synthesis and structure of nanosystems with graphene**

The chapter presents methods for synthesizing graphene on both metal and SiC substrates, with and without intercalation of metals like Co and Pt. The author demonstrates the formation of quasi-free-standing graphene and describes its atomic and electronic structures, including a new $Pt_5Gd$ alloy synthesized beneath graphene.

This chapter is cohesive and engaging, and the results are clearly presented. One point of clarification concerns Fig. 5.20(c): the author might have commented on the absence of visible intensity from the σ-states of graphene in the ARPES image. Additionally, it would be useful to note that the slab band structure calculations appear to have been unfolded along the in-plane direction and stayed unfolded out-of-plane, which may help better understand the calculations.

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**Chapter 6 – Giant Rashba effect in graphene**

Experimental and theoretical results are combined to show that graphene interfaced with heavy metals such as Au and Pt exhibits a giant Rashba-type spin splitting. The author also proposes and characterizes a graphene-based prototype device for spin-orbit-torque magnetoresistive memory applications.

The chapter presents several exciting results. However, a few clarifications and structural adjustments could improve its presentation. For example, the spectral intensity in Fig. 6.3(c) should not be directly compared to the surface states of clean Pt(111), as the Pt concentration in this case is sub-monolayer—an essential point of the “hat” model of giant spin-orbit splitting in Gr/Pt/SiC proposed in the dissertation. Regarding textual consistency, Section 6.3.2 introduces Au intercalation without any prior mention, whereas the earlier focus was clearly on the Gr/Pt/SiC system. A comparison of the functional differences between Pt and Au intercalation layers would be helpful. Also, it might have been more natural to place the description of the graphene-based spin-orbit-torque magnetoresistive memory (SOT-MRAM) after the relevant spectroscopic studies, to preserve logical flow from physical mechanisms to proposed applications.

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**Chapter 7 – Magnetic proximity effect in graphene**

This chapter investigates magneto-spin-orbit graphene with p- and n-type doping, depending on the intercalated species. The author unfolds an enticing story of the interplay between spin-orbit and magnetic exchange interactions. This interplay leads to ferromagnetic and ferrimagnetic ordering in the studied Gr/Au/Co(100) systems, arising from different configurations of interfacial Au and Co atoms.

Notably, these structural differences not only switch the graphene doping from p-type to n-type, but more importantly, they modify the spin asymmetry, resulting in distinct spin textures at the K and K' points, as revealed by spin-resolved ARPES. This study culminates in proposing a model device for detecting circularly polarized infrared radiation based on this effect.

This chapter is an outstanding piece of science, where new physics emerges from experimental results in close synergy with sophisticated theoretical analysis. I would only like to comment that the spin character of electronic states can be unambiguously determined only through ARPES measurements at multiple photon energies. This is because photoelectron polarization reflects not only the intrinsic spin texture of the initial states but also the photon-energy-dependent photoemission matrix elements. However, I fully appreciate that such measurements require synchrotron radiation and substantial beamtime, which justifies that many results in the dissertation are based on data acquired at a single photon energy. Finally, I would point out a few minor issues with cohesiveness, for example, that Section 7.1.1 substantially overlaps with prior Section 6.3.2.

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**Conclusion**

The dissertation concludes by emphasizing the successful engineering of systems that combine spin-orbit and magnetic exchange interactions in low-dimensional materials. The findings provide a solid foundation for developing functional spintronic devices based on graphene and related systems.

Overall, I am impressed by the amount and depth of the new scientific knowledge on the physics of graphene-based two-dimensional systems achieved in this dissertation work. My critique remarks can no way affect my overall assessment of the dissertation as an excellent piece of scientific research. The experimental results are novel, extensive and reliable, and the interpretations are convincing. The dissertation is written in a clear scientific language, comprehensive and a pleasure to read. The main results of the dissertation are reported in 23 publications in refereed journals. Based on these results, three patents are filed. The dissertation stands as a significant development in the field of condensed-matter physics, forming a new scientific direction in the field of spintronics promising significant technological advance. Artem G. Rybkin the author of the dissertation work undoubtedly deserves awarding him the degree of doctor of physico-mathematical sciences on the scientific specialisation 1.3.8 (condensed matter physics).

Yours sincerely

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