Summary of Professional Accomplishments

1. Name

Mateusz Goryca

2. Diplomas, degrees conferred in specific areas of science or arts, including the name of the institution which conferred the degree, year of degree conferment, title of the PhD dissertation

2012	Ph.D. degree in Solid State Physics received with distinction from University of Warsaw, Poland and Université Joseph Fourier, Grenoble, France (now Université Grenoble Alpes) – the Ph.D. work was carried out at both universities within the *cotutelle* program; title of the Ph.D. dissertation: *Spin dynamics in low-dimensional semiconductor structures.*
2007	M.S. diploma in Solid State Physics, graduation with honors; University of Warsaw, Poland.

3. Information on employment in research institutes or faculties/departments or school of arts

11.2020–present	University of Warsaw, Faculty of Physics, Poland – research work and teaching as an assistant professor (PL: *adiunkt*);
09.2017–10.2020	Los Alamos National Laboratory, USA – research work as a Director's Postdoctoral Fellow;
11.2013–09.2017	University of Warsaw, Faculty of Physics, Poland – research work and teaching as an assistant professor (PL: *adiunkt*);
09.2013–10.2013	French National Centre for Scientific Research (CNRS), Grenoble, France – research work as a Visiting Research Associate;
03.2012–08.2013	University of Warsaw, Faculty of Physics, Poland – research work and teaching as an assistant professor (PL: *adiunkt*).

4. Description of the achievements, set out in art. 219 para 1 point 2 of the Act.

The achievement described herein is a cycle of scientific articles on optical studies of two-dimensional spin systems using non-conventional experimental methods.

I. Introduction

In solid state physics, optical experimental techniques play a pivotal role in unraveling the intricate properties of condensed matter systems. By harnessing the light-matter interactions to probe electronic or structural properties, they facilitate the precise determination of optical constants, band structure, and other parameters essential for the design and optimization of novel materials and devices. Among conventional, most commonly used techniques are simple linear time-integrated spectroscopy (including photoluminescence, absorption, Raman scattering, and Fourier transform infrared spectroscopy), time-resolved techniques (including pump-probe spectroscopy), and magneto-optical experiments. However, sometimes to gain access to crucial information on the studied system one needs to resolve to a less conventional approach. This is either due to the need for extreme experimental conditions (*e.g.* extreme pressure or magnetic field, ultra-low temperatures) or entirely different procedures, often borrowed from seemingly unrelated fields of research.

In my work presented here, I exploited two such non-conventional experimental techniques to probe the fundamental properties of two different two-dimensional (2D) spin systems: Transition Metal Dichalcogenide (TMD) monolayers and Artificial Spin Ices (ASIs). First, I used extreme magnetic fields (produced with the world-strongest non-destructive pulsed magnet, among others) to study the fundamental optoelectronic parameters of the TMD family, as well as interesting properties of 2D gas of resident electrons and holes in those materials. Then, I utilized a rare, yet very powerful, technique of optical noise spectroscopy to reveal the dynamic properties of those 2D carrier gases. Finally, employing the know-how gathered in those experiments, I used noise spectroscopy to study a number of interesting phenomena in several ASI systems.

Below, I start with a description of the two studied systems, followed by a discussion of the presented cycle of scientific articles.

I.1. Transition Metal Dichalcogenides

TMDs, such as MoS_2 and WSe_2, are a family of 2D materials widely investigated nowadays due to their quite unusual properties. Their atomically-thin layers have a graphene-like hexagonal crystal structure but – in marked contrast to graphene – they are semiconducting. In their bulk form, TMDs exhibit an indirect band gap [1,2], but they become direct-gap semiconductors when thinned down to a single monolayer [3,4]. This makes them immediately useful for optoelectronics applications such as light emitters and photodetectors. Furthermore, due to the absence of a lattice inversion center, their band structure has two non-equivalent valleys at the K and K' points of the hexagonal Brillouin zone [5,6]. Unlike in most conventional semiconductors, this valley degree of freedom is directly coupled to specific circular polarizations of the optical transitions in these materials. Such valley-specific selection rules arise due to so-called spin-valley locking [5] caused by strong spin-orbit coupling. These two properties of TMDs – the presence of nonzero direct bandgap and the valley degree of freedom directly coupled to light – have rejuvenated interests in the field of *valleytronics*. It is focused on the long-standing idea of exploiting an electron's or hole's valley to encode information [7-12], in the case of TMDs enriched with the presence of an all-optical interface to induce and detect the valley polarization (in marked contrast to most conventional semiconductors).

Last but not least, TMDs (together with some other layered materials) also offer a unique possibility to combine different systems by stacking their monolayers mechanically on top of each other in a designed, precise sequence [13]. This gives an unprecedented fine construction control over the resulting so-called van der Waals heterostructures with properties not found in nature and impossible to create with other methods.

I.2. Artificial Spin Ice

Artificial Spin Ices (ASIs) are two-dimensional metamaterials made of interacting nanomagnets arranged into periodic lattices, wherein the interactions between individual elements can be engineered by the size, spacing, and overall geometry of the lattice [14,15]. Originally they were conceived to emulate the frustrated magnetic interactions in natural pyrochlore spin ice materials (such as $Ho_2Ti_2O_7$ or $Dy_2Ti_2O_7$) which exhibit low-temperature states characterized by a residual entropy, closely related to that of crystalline water ice – hence the name *spin ice*. However, the essentially unlimited freedom to design ASI lattices has soon allowed studies of novel magnetic topologies not found in natural systems and in which the degrees of magnetic frustration, extensive degeneracy, and residual entropy can be intentionally engineered [16,17]. At the same time, the small size and shape anisotropy of each nanomagnet constituting the ASI lattice make it behave – with a very good approximation – as a single Ising-like macrospin, rendering the description and modeling of the ASI system quite straightforward.

One of the most exciting properties of both natural spin ices and ASIs is that the fundamental excitations in those systems have an emergent description in terms of *effective* magnetic monopoles [18-20] – that is, mobile quasiparticles that possess the equivalent of net magnetic charge. These charge excitations originate from a discrete analog of a non-zero divergence of the magnetic field in the ASI lattice. In an archetypal square ASI, each vertex of the lattice consists of four magnetic moments, with nearest neighbors perpendicular to each other. In the lowest-energy state, the orientation of those moments at each vertex obeys the "2-in/2-out" ice rule and thus the vertices possess no magnetic charge. However, a spin-flip of any of the four moments at a given vertex leads to a higher-energy "3-in/1-out" or "1-in/3-out" configuration which can be described with a language of magnetic monopole quasiparticle. Such quasiparticles can interact with each other and with applied magnetic fields via the magnetic analog of the electronic Coulomb interaction, leading to the emergence of a range of novel phenomena [15,21], including the possibility of realization of the idea of "magnetricity" (magnetic analog of electricity) [21].

II. TMD spectroscopy in extreme magnetic fields

From the point of view of TMDs' applications in optoelectronics, properties related to the excitons, the fundamental quasiparticle excitations by light, are particularly relevant. The exciton's mass, size, binding energy, and lifetime are key variables, as well as the dielectric screening properties and the free particle bandgap of the material itself. These material parameters constitute essential ingredients for realistic optoelectronic device models, as well as for the design and engineering of functional van der Waals heterostructures. However, until very recently, many of those fundamental parameters – the exciton masses and the effective dielectric screening lengths in particular – were still assumed from density functional theory calculations [22-24] and have not been experimentally measured.

In principle, these crucial material properties can be directly accessed with the use of optical spectroscopy of excitons in large magnetic fields – and historically has been in many "conventional" III-V and II-VI semiconductors. This is because of how magnetic fields influence exciton energies [25,26]. At field B weak enough to make the cyclotron energy $\hbar\omega_c = \hbar eB/m_r$ significantly less than the exciton binding energy, the lowest-order correction to the exciton's energy is the quadratic diamagnetic shift: $\Delta E = \sigma B^2$, where the diamagnetic coefficient σ depends on the size of the exciton's envelope wavefunction in the plane normal to B, and the exciton's reduced mass m_r. The former depends primarily on the dielectric properties of the material (and its immediate surroundings in the case of low-dimensional structures where the exciton's wavefunction extends beyond the structure itself) [27,28]. In the opposite limit of a strong magnetic field where the cyclotron energy is significantly larger than the exciton binding energy, the latter is negligible compared to the separation between the electron or hole Landau levels (LLs). Under such conditions, interband optical transitions occur between the linearly dispersing LLs in the valence and conduction bands. Thus the transition energies of the excitonic states increase approximately linearly with B, with the slope of $(N+½)\hbar\omega_c = (N+½)\hbar eB/m_r$ (where N is the index of the LLs involved in the transition), if we neglect the spin effects. As such, measuring the linear magnetic field dependence of the excitonic transitions' energies in the regime of strong magnetic fields provides a powerful tool to directly measure the exciton's mass, independent of any other material parameter or model of the electrostatic potential. As mentioned above, such an approach has been successfully used for a plethora of conventional semiconductor systems [29-31], for which the field required for the strong field regime is typically a few tesla. However, the situation with monolayer TMDs is much more difficult. Due to much larger exciton binding energy (hundreds of meVs *vs.* ~5 meV for GaAs) and much smaller exciton radius (~1 nm *vs.* ~10 nm for GaAs), the field required to drive the monolayer TMD into the high field regime is of the order of 1000 T, rendering the experiment impossible.

To solve this issue, in work [**MG1**] we used a twofold approach: first, we focused not only on most strongly bound exciton ground (*1s*) state but also – thanks to the high quality of studied materials and the use of strong magnetic fields – we were able to follow much less-strongly bound excited (*2s*, *3s*, ...) Rydberg states, as in [32] for WSe_2 monolayer. This allowed us to reduce the required magnetic field below 100 T which is still an extreme field, nevertheless experimentally achievable. By using the world-strongest non-destructive pulse magnet [33] we then observed the evolution of the excitonic states visible in the absorption spectra for the entire family of the Mo- and W-based TMDs. From the almost-linear diamagnetic shift of the highest observed Rydberg states alone, we were able to precisely estimate the exciton reduced mass for all those materials without any assumptions on any other material parameters or model of the electrostatic potential. Even more precise estimates of m_r were achieved by modeling the field dependence of all the exciton energies with the use of the popular Rytova–Keldysh potential [34,35]. This also allowed the determination of a number of other crucial material parameters including the exciton binding energies, the free-particle bandgaps, the dielectric screening parameters, and the excitons' sizes. Since our experiments were performed for the entire family of Mo- and W-based TMDs, we were able to experimentally confirm long-standing expectations of heavier exciton masses and larger dielectric screening lengths as the chalcogen atomic mass increases (from S to Se to Te), and also as the metal atom becomes lighter (from W to Mo). Importantly, measured exciton reduced masses turned out to be significantly larger than those predicted by DFT calculations [22-24], especially for Mo-based TMDs, which is consistent with surprisingly heavy electron masses revealed by transport measurements [36,37].

My contribution to the work presented in [**MG1**] was leading and included the design and preparation of unique elements of the experimental setup, carrying out the optical measurements in extreme magnetic fields in collaboration with the second, third, and eighth authors of the paper, performing data analysis together with the second author, fitting the Rytova–Keldysh model to the experimental data to determine key parameters of studied materials, and preparation of the manuscript together with second and the last author.

In follow-up works [**MG3, MG5**] we took advantage of heavy carrier masses determined in [**MG1**], as well as extreme 2D quantum confinement and reduced dielectric screening, that together render monolayer TMDs excellent platform to study electron-electron interactions and many-body correlations. By using similar experimental techniques – absorption spectroscopy in extreme pulsed magnetic fields – we studied in detail *electrostatically gated* TMD monolayers which offered a unique possibility to simultaneously tune both the magnetic field and the carrier density. Many previous optical studies have shown that doping a TMD monolayer with a low density of additional carriers leads to rapid suppression of the neutral exciton absorption resonance (X^0) and the appearance of positively or negatively charged excitons ($X^{\pm}$ trions) at lower energy [38-41]. In the simplest picture, trions are quasiparticles comprising a photoexcited neutral exciton dressed by a resident hole or electron. Some researchers have also proposed a complementary description, valid especially in the regime of small carrier densities, as the attractive branch of exciton-polaron resonance arising from the collective response of the Fermi sea to the photoexcited exciton [42,43]. Regardless of the description, charged excitonic transitions provide a convenient optical means to study the rich physics of the 2D gas of resident carriers in a TMD monolayer.

In [**MG3**], using a hole-doped WSe_2 monolayer, we first demonstrated that the valley Zeeman effect of the resident holes (described with effective hole g-factor g_h) can be tuned with the carrier density in a wide range. Such an observation was possible through optical identification of the number of filled LLs in K and K' valleys separately for any given carrier density, exploiting valley-specific optical selection rules. It is consistent with the expectation that electron-electron interactions increase at small carrier densities leading to enhancement of g_h [44-46], and was crucial to our main goal of controlling the LL alignment in the K and K' valleys. In the case of "conventional" 2D electron or hole gases, such as those in Si, GaAs, or AlAs semiconductors, the control over the alignment of LLs with different quantum numbers is typically achieved by the application of a *tilted* magnetic field which enables tuning orbital (cyclotron) and spin (Zeeman) energies independently. However, in TMD monolayers, due to very strong spin-orbit coupling, spins are locked out-of-plane [46-48], rendering the tilted-B method useless.

Therefore in our work, we exploited the carrier-density-tunable g_h to energetically align chosen LLs in the K and K' valleys and observed that under such conditions the 2D hole gas becomes unstable. As evidenced by the sudden disappearance and reappearance of optical absorption (charged exciton X^+) related to holes in one valley, those holes spontaneously transfer to the opposite valley. This means spontaneous valley polarization, analogous to the transition to the quantum Hall ferromagnet state in conventional semiconductors [49-53]. Whether such instability represents a true first-order phase transition remains an open question, but detailed transport studies in similar fields and doping regimes could answer this question. Particularly, the presence or absence of hysteretic resistance [52-53] could dispel the doubts. Nevertheless, the observed abrupt changes in the 2D hole gas valley polarization highlight the key role of e-e interactions in monolayer TMDs.

In turn, in [**MG5**] we focused on the important role played by many-body correlations in high density *electron* gas in a WSe_2 monolayer. As it has been reported in several previous studies, apart from neutral exciton visible at zero carrier density and charged excitons appearing at low density of additional holes or electrons, an additional strong absorption feature emerges in high-quality WSe_2 monolayers in the regime of large electron densities [38,39,54-57]. The origin of this resonance (often referred to as $X^{-\prime}$), located well below the energy of any trion or attractive polaron resonance, remained unclear while various interpretations have been proposed: from a fine structure of the X^- trion, to a doubly charged negative trion, to a new quasiparticle arising from exciton interactions with intervalley plasmons [39,55,56,58]. First optical studies in the magnetic field have shown, however, that the resonance in question evolves into a series of discrete absorption peaks in the applied field, which was interpreted remarkably well within a simple picture of single-particle optical transitions between LLs in the valence and conduction bands [55]. Such an interpretation was somewhat surprising, given the very large exciton binding energies and strong e-e interactions in TMD monolayers.

Therefore, to shed more light on the origin of the X' resonance, we performed absorption spectroscopy studies of a WSe_2 monolayer in the regime of strong electron doping, in magnetic fields almost an order of magnitude stronger than those used in [55]. The use of such strong fields allowed us to clearly demonstrate repeated filling and emptying of the lowest (0th) optically active LL in the K' valley by the dense electron sea during the field sweep, evidenced by multiple quenchings and reappearances of the corresponding optical transition due to Pauli blocking. Such observation remains consistent with earlier studies [55] and with the single-particle picture. Crucially, however, we also showed that these quenchings and reappearances are accompanied by changes in the energy and oscillator strength of optical absorption to higher-lying LLs in both the K' and K valleys, which are not occupied. Correspondence between those optical transitions unambiguously demonstrates that the X' resonance is a many-body state with intervalley correlations, *i.e.* it can be described as a state involving not only the photoexcited electron-hole pair but also opposite-spin electrons in the same valley, as well as both up- and down-spin electrons in the opposite valley. In [**MG5**] we did not elaborate on the exact composition of the X' wavefunction, however a later theoretical work by D. Van Tuan et al. [59] has shown that our findings can be successfully described with a language of a six- and eight-body exciton states ("hexcitons" and "oxcitons", respectively), depending on the number of distinguishable electron reservoirs with which the photoexcited e-h pair can interact.

In the case of [**MG3**] and [**MG5**], my contribution was significant and included performing the measurements in extreme magnetic fields (together with the first and fourth author in [**MG3**] or together with the last author in [**MG5**]), performing data analysis together with the first author, and preparation of the manuscript together with the first and the last author.

The role of many-body interactions in enhancing the electron magnetic susceptibility in other TMD monolayers was also the central focus of my later work [60], beyond the scope of the achievement presented herein.

III. Revealing TMD valley relaxation times with valley noise detection

After establishing the fundamental optoelectronic properties of the TMD monolayers with the use of extreme magnetic fields, we continued our studies with a focus shifted toward the dynamic properties. Of particular interest was one of the flagship properties of those materials: the ability to optically encode information in electron's valley degree of freedom, which forms the conceptual basis of the field of valleytronics. Naturally, for any valley-based information processing scheme, the intrinsic time scales of valley relaxation are critically important, which constituted our main motivation for the studies presented in [**MG2**]. Many prior studies using the standard pump-probe approach have demonstrated that while valley-polarized excitons scatter very quickly on picosecond time scales [61-64], the valley relaxation of *resident* electrons or holes can be orders of magnitude longer [65,66], up to the microseconds range in the case of holes at low temperatures [67]. However, the issue with all those pump-probe studies is that, by definition, they are *perturbative*. A strong optical pump pulse is used to inject nonequilibrium electrons and holes that scatter, dissipate energy, and interact, driving the valley polarization of the resident carriers away from thermal equilibrium. For one, the nature of such a process is not fully understood yet, but more importantly, optical pumping of both photoexcited electrons and holes can create optically inactive "dark" excitons and trions [68-70] whose lifetime is typically many orders of magnitude longer than their optically active counterparts'. The presence of such a long-lived reservoir of spin-and-valley polarized excitons could, in principle, mask the detection of resident carriers' valley relaxation, as suggested by some researchers [71].

For those reasons, in our studies we used a non-standard approach: optical noise spectroscopy. Per the fluctuation-dissipation theorem [72], there is a fundamental relationship between any system's dynamic linear response and its intrinsic thermal fluctuations under conditions of thermal equilibrium: the temporal correlation function of the thermal fluctuations is determined by the intrinsic time scales of scattering and relaxation, which is what is measured in a typical pump-probe experiment. Thus, both approaches (pump-probe and noise-based) provide essentially the same information. The difference is that the noise-based experiment does not require any perturbation of the system and thus avoids problems related to strong perturbation or dark excitons.

In studies presented in [**MG2**], we used an electrostatically gated, hole-doped WSe_2 monolayer deposited on a transparent substrate, to allow transmission of light through the sample. Due to valley-selective optical selection rules, the absorption of left and right circularly polarized light ($\sigma^{\pm}$) at energies near the positively charged exciton resonance strongly depends on resident holes densities $p^{\pm}$ in K and K' valley. Associated with the $\sigma^{\pm}$ absorptions are the dispersive indices of refraction $n^{\pm}(E)$, the difference between which is directly measured by optical Faraday rotation (in the case of light transmission through the sample). Therefore, the Faraday rotation (and closely related Kerr rotation for the reflectance configuration) is fundamentally sensitive to the difference between holes density in the opposite valleys: $p^{+} - p^{-}$, *i.e.* the valley polarization, and has been frequently used as a probe of nonequilibrium valley polarizations in standard pump-probe studies [66,67,71]. In the situation without a pump pulse, that is under conditions of thermal equilibrium, the time-averaged hole valley polarization is strictly zero, and therefore no average Faraday rotation is observed. However, in any real system at finite temperature, thermodynamic fluctuations exist and so holes in the WSe_2 monolayer can spontaneously scatter between K and K' valleys, leading to a small valley polarization that fluctuates randomly in time about zero.

In our work we have shown that this "valley noise" can indeed be detected as tiny fluctuations of Faraday rotation, analogously to studies of optical *spin* noise spectroscopy in atomic alkali vapors [73] and some conventional semiconducting systems [74,75]. From a narrow Lorentzian power spectrum of those fluctuations, equivalent to the Fourier transform of the temporal correlation function [72], we deduced a very long valley relaxation time (of the order of half a microsecond) of the holes at low temperatures, and about an order of magnitude shorter one for electrons – in agreement with the expectation that spin-valley locking is much weaker in the conduction bands than in the valence bands of monolayer TMDs [76].

An important point of our work was to resolve long-lasting concerns about the earlier pump-probe studies of doped TMD monolayers regarding the long-lived dark states or other perturbations that can affect the slow valley relaxation of resident carriers. To resolve those concerns, we directly compared the results of the noise-based experiments to the valley relaxation measured independently by optical pump-probe experiment using the same WSe_2 monolayers. An excellent agreement that we obtained validates the interpretation of the conventional perturbative pump-probe approach. Importantly, such validation would not be possible without perturbation-free measurements. The fluctuation-based method is particularly well suited for systems with exceptionally slow relaxation where the intrinsic relaxation time scales revealed by noise can be longer than the time scales accessible with perturbative methods. For such systems, any experimentally related perturbation may be stronger than the very weak interactions that lead to slow relaxation.

My contribution to **[MG2]** was leading and included the design and preparation of the experimental setup, performing the optical measurements with help from the third author, performing all data analysis, and preparing the manuscript together with the last author.

IV. Magnetic monopoles in ASIs studied via noise spectroscopy

At the time of the experiments described above, we were also studying a different type of a 2D spin system: a square ASI. While many previous studies have established a number of interesting static properties of that system, most notably the presence of magnetic monopoles [18-20], the dynamical studies of the system kinetics were much less developed. This was mainly due to the technical limitations of magnetic imaging techniques most commonly used to probe ASIs [77-79], and the existing studies of system kinetics were limited to a rather narrow range of time scales and characteristic frequencies [80-82]. Additionally, while external magnetic fields are quite obvious tuning parameter for ASIs (which by definition are magnetic systems), most of the abovementioned techniques are either difficult or simply impossible to combine with applied fields. For that reasons, an alternative means of accessing the ASI dynamics were highly desired.

Having established a powerful noise-based optical toolset for studies of spin-and-valley dynamics in TMDs, we quickly realized that an analogous approach can prove extremely useful in the case of ASIs, circumventing the limitations of most commonly used standard experimental techniques. The frequency range covered by a noise spectrometer can, in principle, span over many orders of magnitude, while our all-optical approach is perfectly compatible with applied magnetic fields. Therefore we followed this idea by establishing a high-bandwidth magneto-optical noise spectrometer to passively detect spontaneous magnetization fluctuations in square ASI **[MG4]**. A weak linearly polarized probe laser was focused on and reflected from the ASI surface at a large incidence angle. Due to the longitudinal magneto-optical Kerr effect, magnetization fluctuations on the sample surface imparted Kerr rotation fluctuations on the polarization of the reflected laser, which were detected with balanced photodiodes. The frequency spectrum of those fluctuations was recorded in a wide frequency range (typically spanning 5-6 orders of magnitude), allowing for detailed analysis of underlying spin dynamics. Thanks to the virtually unlimited ability to design the geometry of the nanoislands constituting the ASI lattice, they were prepared small and thin enough to make them thermally active near room temperature, meaning that in the absence of biasing fields, each island's spin can flip back and forth spontaneously. These spontaneous flips ensure that the lattice can efficiently sample the vast manifold of possible moment configurations and remain near its magnetic ground state in thermal equilibrium.

The main result of **[MG4]** was the discovery that the square ASI lattice can host, in specific regions of its magnetic phase diagram, plasma-like regimes containing a high density of mobile magnetic monopoles. These regimes result from the vanishing tension on the Dirac strings connecting mobile monopoles at the crossovers between different magnetic orderings of the system. At such crossovers, certain spin flips – and associated motion of monopoles – have no energy cost, allowing the monopoles to diffuse freely through the lattice. Detailed analysis of the frequency spectrum of the magnetization fluctuations – and thus underlying monopole kinetics – revealed that monopoles' motion is most diffusive (*i.e.* minimally correlated) in the plasma regime. The experimental results were strongly supported by detailed Monte Carlo simulations which allowed the observation that both the density of monopoles and their kinetic properties can be tuned with the magnetic field. Altogether, those results opened the door to new exotic regimes of effective magnetic charges in synthetic matter and provided a new framework for probing their physics.

My contribution to the work presented in [**MG4**] was leading and included the design, construction, and optimization of the broadband noise spectrometer, carrying out all the experimental work, analyzing the data in collaboration with the last author, performing the key Monte Carlo simulations (preliminary simulations were also performed by the seventh author), and preparation of the manuscript, together with the last author.

In the follow-up work [**MG6**] we used the same research framework to study further details of the square ASI as well as the recently introduced quadrupolar ASI [83]. Of particular interest were the magnetic field-dependent phase diagrams and phase transitions of those systems. With the use of magnetization noise spectroscopy, we identified stable phases in the magnetic field and regimes of extensive degeneracy, while Monte Carlo simulations delivered details of the magnetic-field-dependent equilibrium thermodynamic properties (*e.g.* specific heat and magnetic order parameters). This allowed us to fully reconstruct field-dependent phase diagrams and reveal a rich diversity of magnetic orderings and phase transitions that can occur in studied ASI systems. In particular, we analyzed in detail two exotic phases in quadrupolar ASI: ferro-quadrupolar and antiferro-quadrupolar (first reported in [83]) and demonstrated, that the latter one is much less stable. Similarly to the square ASI, at the boundaries between the field ranges characterized with stable long-range magnetic order, the quadrupolar ASI also exhibits regimes where topologically protected, monopole-like magnetic excitations can proliferate and diffuse. With the help of Monte Carlo simulations, we analyzed the nature of those excitations and their similarities to the magnetic monopoles observed in square ASI.

The Monte Carlo simulations played an even more profound role in [**MG7**] where we studied an ASI system known as "tetris ice" [84,85]. Formed by the selective removal of moments from the canonical square ice lattice, tetris ASI belongs to a group of vertex-frustrated ASIs [86] in which not every lattice vertex can have its moments arranged in its local low-energy configuration at the same time. This leads to many intriguing phenomena, including extensive degeneracy of the ground state and long-range ordering driven by the *increase* of the entropy [87] (while in classic physical systems, such ordering is associated with a *decrease* in entropy). However, despite exhibiting such rich physics in the absence of an external magnetic field, tetris ASI has not been studied in applied fields prior to our work. Thus, we studied the field-dependent behavior of tetris lattice, again by combining the optical detection of its intrinsic magnetization noise with detailed Monte Carlo simulations. The experimental results revealed particularly intense and narrow bands of noise for certain directions and ranges of the applied field. Due to the complex nature of those noise signatures and the non-trivial geometry of the tetris lattice, Monte Carlo simulations were indispensable to deconstruct these noise features and tease apart the fluctuations of individual moments in ways that are experimentally intractable. By doing so, we identified the noise signatures in question as arising from a collective behavior of magnetic excitations which can fractionalize and separate into two quasiparticles that can diffuse along extended quasi-1D spin chains in the lattice. These results again demonstrated the power of noise-based studies and sophisticated Monte Carlo simulations to reveal microscopic details of complex magnetic phenomena.

My contribution to both [**MG6**] and [**MG7**] was leading and included conceiving the research together with key co-authors, designing, constructing, and optimizating of the experimental setup, performing all optical measurements, data analysis, and Monte Carlo simulations, as well as preparation of the manuscript together with the last two authors of the articles.

V. Summary

The results discussed here are related to two two-dimensional spin systems: TMD monolayers and ASI lattices, the studies of which are young, extremely active and quickly developing fields in condensed matter physics. In my research, I used unconventional, rarely utilized experimental techniques: magnetic fields of the order of 100 T and non-perturbative optical noise spectroscopy. This enabled observation of a number of important and intriguing properties of these systems, including the precise determination of the fundamental optical parameters of TMD materials, revealing the spin dynamics of both systems in a way that is free from concerns common for traditional studies, and the discovery and elucidation of interesting collective spin behaviors in both systems. The importance and recognition of my results in the scientific community, as well as their significant contribution to the development of the field to which they relate, is evidenced not only by the presented scientific articles published in prestigious international journals but also by the number of invited talks – the details can be found in the *list of scientific achievements* attached.

VI. References

The cycle of scientific articles constituting the achievement described herein:

[MG1] **M. Goryca**, J. Li, A. V. Stier, T. Taniguchi, K. Watanabe, E. Courtade, S. Shree, C. Robert, B. Urbaszek, X. Marie and S. A. Crooker, "*Revealing exciton masses and dielectric properties of monolayer semiconductors with high magnetic fields*", Nature Comm. **10**, 4172 (2019).

[MG2] **M. Goryca**, N. P. Wilson, P. Dey, X. Xu and S. A. Crooker, "*Detection of thermodynamic "valley noise" in monolayer semiconductors: Access to intrinsic valley relaxation time scales*", Science Advances **5**, eaau4899 (2019).

[MG3] J. Li , **M. Goryca**, N. P. Wilson, A. V. Stier, X. Xu and S. A. Crooker, "*Spontaneous Valley Polarization of Interacting Carriers in a Monolayer Semiconductor*", Physical Review Letters **125**, 147602 (2020).

[MG4] **M. Goryca**, X. Zhang, J. Li, A. L. Balk, J. D. Watts, C. Leighton, C. Nisoli, P. Schiffer and S. A. Crooker, "*Field-Induced Magnetic Monopole Plasma in Artificial Spin Ice*", Physical Review X **11**, 011042 (2021).

[MG5] J. Li, **M. Goryca**, J. Choi, X. Xu and S. A. Crooker, "*Many-body exciton and inter-valley correlations in heavily electron-doped* WSe_2 *monolayers*", *Nano Lett.* **22**, 426 (2022).

[MG6] **M. Goryca**, X. Zhang, J. D. Watts, C. Nisoli, C. Leighton, P. Schiffer, and S. A. Crooker, "*Magnetic field dependent thermodynamic properties of square and quadrupolar artificial spin ice*", Physical Review B **105**, 094406 (2022).

[MG7] **M. Goryca**, X. Zhang, J. Ramberger, J. D. Watts, C. Nisoli, C. Leighton, P. Schiffer, and S. A. Crooker, "*Deconstructing Magnetization Noise: Degeneracies, Phases, and Mobile Fractionalized Excitations in Tetris Artificial Spin Ice*", Proceedings of the National Academy of Sciences, in press (2023).

Other publications:

[1] J. A. Wilson and A. D. Yoffe, *The transition metal dichalcogenides: discussion and interpretation of observed optical, electrical and structural properties*, Adv. Phys. **18**, 193 (1969).

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[3] A. Splendiani, L. Sun, Y. Zhang, T. Li, J. Kim, C.-Y. Chim, G. Galli, and F. Wang, *Emerging Photoluminescence in Monolayer* MoS_2, Nano Lett. **10**, 1271 (2010).

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5. Presentation of significant scientific or artistic activity carried out at more than one university, scientific or cultural institution, especially at foreign institutions

After receiving my master's degree (with honors) in 2007, I began doctoral studies at the Faculty of Physics, University of Warsaw, and at Université Joseph Fourier (now Université Grenoble Alpes) in Grenoble, France. The thesis was carried out at both universities simultaneously, within the framework of the Polish-French *cotutelle* program, under the guidance of two supervisors: a Polish one (Prof. Dr. Piotr Kossacki) and a French one (Prof. Dr. Marek Potemski). For the realization of the part of the Ph.D. work carried out in France, I was awarded a prestigious and highly competitive scholarship by the French government. Experimental research for that part of the work was conducted at the Grenoble High Magnetic Field Laboratory (GHMFL). The work was devoted to the spin dynamics of magnetic Mn^{2+} ions in CdTe low-dimensional structures, with particular emphasis on single quantum dots containing single magnetic ions [MG8-MG10]. During its realization, I was also involved in other scientific projects, primarily devoted to single, undoped CdTe dots [MG11-MG12].

After being awarded my Ph.D. (with honors) in 2012, I was hired as an assistant professor at the Faculty of Physics, University of Warsaw, where I worked until 2017 with a two-month break in 2013. Initially, I continued research related to quantum dots doped [MG13-MG18] and undoped [MG19-MG22] with magnetic ions. In 2013, I took an employment opportunity at the French National Center for Scientific Research (Centre National de la Recherche Scientifique, CNRS) in Grenoble and was hired for a two-month internship as a visiting research associate. My work was devoted to studies of resonant excitation of TMD monolayers, which initiated a new avenue of my research related to this material system. After my internship, I continued my work at the University of Warsaw, devoting more attention to this field [MG23-MG25]. I also conducted research on low-dimensional CdTe structures containing magnetic ions using optically detected magnetic resonance (ODMR) [MG26-MG27].

In 2017, I was awarded a prestigious and highly competitive Los Alamos National Laboratory (LANL) Director's Postdoctoral Fellowship to pursue an internship at the facility, and began a more than three-year term in the group of Dr. Scott Crooker that also involved the studies of TMD monolayers – this time using unconventional experimental techniques: extreme magnetic fields and optical noise spectroscopy. The results of these studies were the subject of publications that are included in the cycle discussed in section 4. above ([MG1-MG3,MG5]). In parallel, using the second of these techniques, I began to study the dynamics of other two-dimensional spin systems: the ASI lattices [MG4]. In addition, I was also involved in the studies of colloidal quantum dots [MG28,MG29], as well as polar and chiral magnetic materials [MG30].

After completing my fellowship at LANL in 2020, I was again employed at the Faculty of Physics, University of Warsaw as an assistant professor and as the PI of a project funded by the National Agency for Academic Exchange within the Polish Returns program. The studies I am currently engaged in are both a continuation of previously undertaken topics: dynamics of the ASI systems [MG6,MG7] and TMD monolayers [MG31], as well as addressing new research fields (low-dimensional perovskite structures).

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6. Presentation of teaching and organizational achievements as well as achievements in popularization of science or art

Supervision over diploma theses

- assistant supervisor of Ph.D. thesis of Julia Kucharek (ongoing), Faculty of Physics, University of Warsaw;
- assistant supervisor of Ph.D. thesis of Tomasz Smoleński, "*Spectroscopy of CdSe/ZnSe quantum dots with individual Fe^{2+} ions*", Faculty of Physics, University of Warsaw, 2018;
- supervisor of M.S. thesis of Melania Deresz, "*Pulsed measurements of magnetization relaxation of diluted magnetic semiconductors*", Faculty of Physics, University of Warsaw, 2014;
- supervisor of M.S. thesis of Małgorzata Zinkiewicz, "*Measurements of the spin relaxation time of Mn^{2+} ion in selenide-based structures*", Faculty of Physics, University of Warsaw, 2014;
- supervisor of B.S. thesis of Tomasz Łopion, "*Optical spectroscopy of vanadium-doped tungsten disulfide monolayers*", Faculty of Physics, University of Warsaw, 2023;

- supervisor of B.S. thesis of Wojciech Kolesiński, "*Fabrication of exfoliated semiconducting structures using dry stamp method*", Faculty of Physics, University of Warsaw, 2022.

Teaching

During my employment at the Faculty of Physics, University of Warsaw, I taught a number of subjects, including:

- *Introduction to optics and condensed matter physics* (lecture and exercises for third-year undergraduate students);
- *Computational tools in the analysis of experimental data of condensed matter physics* (exercises for first-year students of the second degree program; the concept and program of the course was developed by myself);
- *Physical and electronic laboratory* (laboratory for second-year undergraduate students);
- *Measurement uncertainty analysis and introduction to laboratory* (laboratory for second-year undergraduate students);
- *Specialized laboratory* (laboratory for first-year students of the second degree program);
- *Physics with mathematics* (exercises for first-year undergraduate students);
- *Team student projects* (team project for undergraduate students).

Additionally, during the course of my doctoral work, I taught the following classes:

- *Computer lab* (exercises for first-year undergraduate students);
- *Physics with mathematics* (exercises for first-year undergraduate students);

Popularization of science

- From 2010 to the present (with a pause in years 2017-2020 due to a foreign post-doctoral fellowship) I have been a member of the Committee of the Polish Physics Olympiad. In the years 2014-2017, I additionally held the position of Olympiad's Scientific Secretary for Experimental Problems, when I was responsible for the development and organization of the experimental problems of the Olympiad. My current duties include evaluating the participants' solutions to the competition problems, co-authoring the problems, and training the Polish representation for the International Physics Olympiad. I also contribute to the organizational side of the Olympiad.
- In the years 2012-2014, together with Professor of Education Marek Golka, I taught classes for talented young students from the XIII High School in Szczecin within a summer science camps.
- In years 2011-2013, I hosted workshops for middle and high school students as part of the Summer School of Physics, as well as lectures within Open Classes in Physics, at the Faculty of Physics, University of Warsaw.

Organizational contribution

- Since 2021, I have been responsible for overseeing the implementation of the modernization and expansion of the helium gas recovery and liquefaction infrastructure at the Faculty of Physics, University of Warsaw. These activities are carried out within the framework of the *Helium Infrastructure for Ochota Campus* activity, funded within the *Excellence Initiative - Research University* project at the University of Warsaw.
- In the years 2016-2017, I served as a member of the Council of the Faculty of Physics, University of Warsaw.
- In the years 2014-2017, I supervised the Choir of the Faculty of Physics, University of Warsaw.

Mateusz Goryca

(Applicant's signature)