CNRS-Momentum 2018

SCIENTIFIC PROJECT

LAST NAME: MARTIN
First Name: Nicolas

THEME CHOSEN:

10. Showing the invisible

TITLE

A Journey to the Heart of Matter: Unveiling Invisible Properties of Topological Defects at the Nanoscale

RESEARCH TOPICS – KEYWORDS

- **Topic 1:** Condensed matter: organization and dynamics
- **Topic 2:** Condensed matter: structures and electronic properties
- **Keywords:** Topological Defects, Magnetic Skyrmions, Large Scale Research Facilities, Scattering Techniques

ABSTRACT

Condensed matter is an arena of choice where one can observe and manipulate novel forms of quantum objects. By virtue of *chemistry*, it is indeed possible to map *mathematical* concepts into the *physical* world. One prominent example is the so-called *skyrmion* (SK), a particle-like solution appearing in non-linear field theory and actually observed in certain class of magnetic materials. This recent discovery has raised a tremendous activity throughout the world because SKs are seen as promising building blocks for next-generation electronics. On the other hand, their study is relevant to different branches of physics, where *topological defects* play a major role (vortices in superconfluids, flux lines in superconductors, dislocations in liquid crystals, strings in inflation cosmology, *etc.*). According to current models, magnetic skyrmions should be observed in a large variety of conditions but up to now, only a small portion of this landscape has been actually explored. Here, we propose to pave a way across this *terra incognita* by following a *transversal* research program. We will identify the material specificities which allow for the emergence of new types of magnetic SKs. More specifically, we will consider chemical doping, magneto-elastic coupling and magnetic frustration as ways to control the properties of SK-host bulk compounds and of the SKs themselves. These *invisible* features will be studied in the reciprocal space using quantum beam diffraction and spectroscopy (neutrons, X-rays & muons). The experimental results will be analyzed in the light of recent theories and dedicated numerical simulations. Our strategy is based on a *strong interconnection between specialists of different fields*, namely chemists, theoretical and experimental physicists. Gathering their expertise, we anticipate a strong impact in the field. In view of its high ambition and transdisciplinary inspiration, we believe that our project is fully relevant to the *Momentum* work program.

MARTIN Nicolas

Condensed Matter Physicist

Born on 03/17/1984, French nationality
Marital life, 2 children (Emmy, born on 08/31/2014 and Samuel, born on 08/17/2016)
Researcher ID **:** 0000-0002-3255-3326

Scientific positions

- **Since 08/2016 :** Scientist in charge of the small-angle neutron scattering (SANS) instrument PA20 at Laboratoire Léon Brillouin (LLB)
- **11/2014 – 04/2016 :** Postdoctoral fellowship at LLB (funded by LabEx "PALM"), on "*Chiral magnets with itinerant magnetism*"
- **04/2012 – 10/2014 :** Beamline scientist at the neutron resonant spin echo instrument RESEDA at the Maier-Leibnitz Institute (MLZ, Germany)

Education

- **2012 :** Ph.D. in Physics from the Université Joseph Fourier (Grenoble) : "*Etude structurale et dynamique de plusieurs systèmes magnétiques par la technique de l'écho de spin neutronique résonant*", supervised by L.-P. Regnault (CEA Grenoble)
- **2008 :** M.Sc. in Physics from the Université Joseph Fourier (Grenoble), speciality "*Physical Methods and Physical Chemistry*" with honors (mention Bien)

Publication record

- *h*-index = 6 (≈ 100 citations), with a total of 12 regular articles (including 1 Phys. Rev. X, 2 Phys. Rev. Lett. and 7 Phys. Rev. B), plus 6 conference proceedings

Scientific and technical skills

- Expert in *neutron scattering* (triple-axis and high-resolution spectroscopy, diffraction & SANS)
- Solid experience in *X-ray diffraction & spectroscopy* (photoemission, magnetic dichroism) and *muon spin relaxation* (µSR)
- Technical development of the neutron resonant spin echo ZETA option on triple axis spectrometer IN22 (Institut Laue Langevin (ILL), Grenoble)
- Upgrade of the neutron resonant spin echo instrument RESEDA (Maier-Leibnitz Institute, Garching bei München, Germany)
- Co-proposer (for the ILL "Endurance 2" program, 2017) of the SAM project, focused on a new SANS instrument equipped with a "MIEZE" option. *Scientific case:* high-resolution study of the statics & dynamics of nanoscopic structures (helimagnets, skyrmion and vortex lattices, macromolecular complexes, cell membranes, *etc.*)

Education and science management

- Remedial courses for 1st year students at the Université Joseph Fourier
- Supervisor of 2 Masters students at the Technische Universität München
- Laboratory courses on neutron scattering at the MLZ and LLB
- Referee for *Journal of Magnetism and Magnetic Materials* and *Review of Modern Physics*
- Member of the laboratory council at LLB: non-permanent scientists representative

1. State-of-the-art, objectives and proposed work

Introduction: Topology and Magnetism – *Topology* is an important branch of mathematics, which establishes equivalences between *a priori* different objects, these equivalences being based on the shape of these objects. The concept of *topological protection* is a by-product of this approach, related to the existence of some conserved quantities upon continuous deformation, with applications in many fields of science. In *solid-state physics*, the introduction of such ideas for the explanation or prediction of novel phases and phase transitions of matter traces back to the 1970-80ies. Some of these crucial contributions have been acknowledged by the Nobel Prize (awarded to Haldane, Kosterlitz and Thouless in 2016), in view of their relevance to a large class of systems. These include semiconductors, two-dimensional quantum liquids and magnets, spin chains or, more recently, Weyl semimetals and topological insulators. In the last decade, the notion of *topology* has gained a renewed interest in the field of chiral *magnetism* following the prediction and discovery of a novel form of particle-like nanometre-sized spin textures, the so-called *skyrmions* (SK)[1]. Their name is inherited from a mechanism proposed by T. Skyrme[2] to describe solutions of non-linear equations in particle physics. As a solution of the original Skyrme model, one finds stable configurations for the elementary nuclei. They are classified according to an integer invariant, the *topological charge*, thereby explaining their stability (**Fig. 1a**).

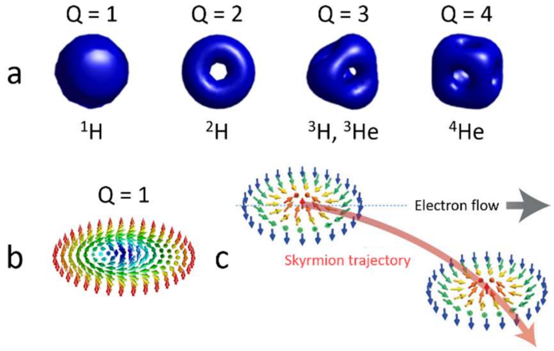

Fig. 1 *– Solutions of the Skyrme model, ordered as a function of increasing topological charge Q*[3]*. Q is connected to* ***a)*** *the symmetry of isoenergy surfaces of light nuclei or* ***b)*** *number of full twists of the local spin vector in the case of magnetism (here 1).* ***c)*** *Current-induced motion of a skyrmion*[4]*.*

In the context of magnetism, SKs are thermodynamically stable *topological defects*, akin to spin vortices (**Fig. 1b**). In bulk materials, they are found in small pockets of the phase diagrams of certain metals and oxides, multiferroic insulators or semi-conducting compounds[5]. These magnetic SKs are topologically equivalent to the magnetic bubbles forming in ferromagnetic thin films but display much smaller characteristic sizes (nm versus µm). They usually carry a topological charge Q = 1 and behave as protected effective magnetic fluxes. As a consequence, these SKs can be manipulated by electric or heat currents, and thus act as movable nanometric information carriers (**Fig. 1c**). These unique features lead to exciting perspectives, especially regarding their use as building blocks for next-generation logic gates or high-density solid-state memories[6].

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[1] A.N. Bogdanov & D.A. Yablonskii, Sov. Phys. JETP **95**, 178 (1989); S. Mühlbauer *et al.*, Science **323**, 915 (2009)

[2] T.H.R. Skyrme, Nuclear Physics **31** (1962) 556-569

[3] Pictures taken from M. Mostovoy, *Multiferroic skyrmions*, 5th Conference on Nuclei and Mesoscopic Physics

[4] Picture taken from G. Chen, Nature Physics **13**, 112 (2017)

[5] X.Z. Yu *et al.*, Nature Mat. **10**, 106 (2011); Y. Tokunaga *et al.*, Nature Comm. **6**, 7638 (2015); A. Nayak *et al.*, Nature **548**, 561 (2017); X.Z. Yu *et al.*, Nature Communications **5**, 3198 (2014); S. Seki *et al.*, Science **336**, 198 (2012) W. Münzer *et al.*, Phys. Rev. B **81**, 041203(R) (2010); I. Kézsmárki *et al.*, Nature Materials **14**, 1116 (2015)

[6] X. Zhang *et al.*, Scientific Reports **5**, 9400 (2015); A. Fert *et al.*, Nature Nanotechnology **8**, 152 (2013)

On the other hand, their study is relevant to different areas of physics, where *topological defects* play a major role (vortices in superconfluids, flux lines in superconductors, dislocations in liquid crystals, strings in inflation cosmology, *etc.*). ***The crucial role of Chirality –*** Magnetic SKs have a *chiral* nature, which means that they differ from their mirror image. In "standard" cases, their existence is due to spin-orbit coupling (SOC), as allowed in systems where inversion symmetry is broken (*i.e.* non-centrosymmetric crystal structure in bulk systems or at interfaces in thin films and multilayers). SOC is characterized by a parameter *D*, acting as a perturbation of the dominant ferromagnetic (FM) exchange *J* and the periodicity λ of the resulting helical order is then controlled by the ratio *J/D*. Under an applied magnetic field and *at* temperatures close to the paramagnetic transition line (red area in **Fig. 2a**), spin helices (**Fig. 2b**) reorient perpendicular to the field and combine to form SKs with typical size λ, sitting on the vertices of a hexagonal lattice (**Fig. 2c**).

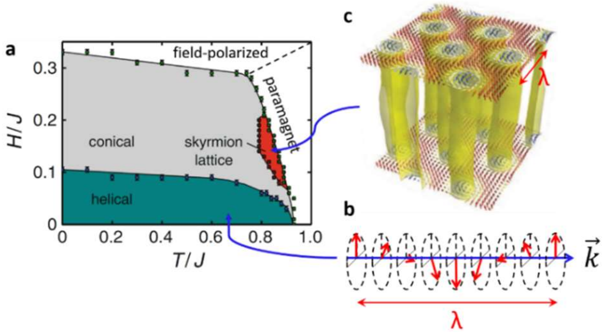

***Fig. 2** – **a)** Generic phase diagram of usual non-centrosymmetric SK-forming systems. **b)** Helimagnetic ground state favoured by the competition between FM exchange and SOC. **c)** Under an applied-magnetic field and close to the critical temperature, a SK lattice[7] can be observed in a small (H,T)-pocket (red area in **a**).*

Beyond this well-established paradigm, recent theoretical works show the possibility to obtain SKs *in systems lacking SOC*[8]. Such studies consider triangular magnets with a strong uniaxial anisotropy and a competition between FM nearest-neighbour (NN) and anti-FM (AFM) next-NN (NNN) exchange interactions. *At low temperature*, these ingredients lead to spin *chirality* and a plethora of multiply twisted states, including a SK lattice (**Fig. 3**)[9].

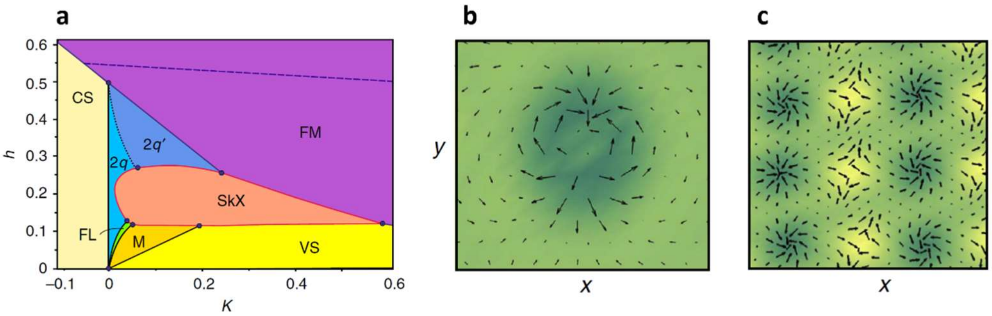

***Fig. 3** – **a)** Typical low temperature phase diagram of a triangular lattice antiferromagnet (TLA). The SK lattice (SkX) is obtained at intermediate values of anisotropy K and magnetic field h. **b)** Metastable SK with topological charge Q = 2 stabilized in a TLA. **c)** SK/anti-SK lattice as stabilized in a TLA.*

These SK states have not been observed yet, but they deserve interest for at least four reasons:

- They are built upon interactions whose length scale correspond to chemical lattice constants (a few Å). As a consequence, they can be potentially *much smaller* than the SOC-based SKs (a few nm).

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[7] Figure taken from P. Milde *et al.*, Science **340**, 1076 (2013)

[8] T. Okubo *et al.*, Phys. Rev. Lett. **108**, 017206 (2012)

[9] A. Leonov & M. Mostovoy, Nature Comm. **6**, 8275 (2015)

- These SKs can display attractive interactions. This permits their clustering and the emergence of bound states with topological charges differing from $Q = 1$ (**Fig. 3b**).
- Since chirality is induced by symmetric exchange interactions, SKs can display distinct helicities, *i.e.* clockwise or anticlockwise twist of the magnetization (**Fig. 3c**). This offers yet additional degrees of freedom for their manipulation.
- The inversion symmetry breaking introduced by the presence of these SKs can lead to a finite electric polarization, hence to multiferroic properties.

Tuning the properties of SKs – The preceding theoretical aspects illustrate the richness of the *skyrmion* concept. In order to go one step further and explore the flexibility of magnetic SKs, it is desirable to look for systems where the above mechanisms (SOC *and* competition between FM and AFM interactions) contribute simultaneously. To that end, one can take advantage of studies on *bulk compounds*, which allow tailoring the magnetic environment by acting on the *chemical* and *physical* properties of the studied samples:

i. Chemical doping – Chemical substitution of magnetic ions is an efficient way to modify the main interactions (J, D) and anisotropies. Low doping usually leads to marginal modifications of the magnetic order (helical periodicity and ordering temperature). At a higher level, the magnetic structure can be strongly destabilized and eventually vanish at a quantum critical point. In between these two extremes, there exist large varieties of situations which can be exploited to tune the magnetic properties of SK-host compounds and of the SKs themselves (size and density).

ii. Magneto-Elastic Coupling – Applying a mechanical stress is an instrumental tool to control the properties of SKs[10]. In fact, most SK hosting systems display pronounced *magneto-elastic* anomalies[11]. These effects can be specifically studied by the application of external constraints, such as uniaxial strain or hydrostatic pressure.

iii. Magnetic frustration – Magnetic *frustration* denotes the impossibility to simultaneously satisfy all magnetic interactions. This usually leads to a reduction of magnetic ordering temperature and allows for the control of the temperature range within which SKs can occur.

2. Methodology and experimental approach

Outline of the Research Program – As shown in the previous section, bulk magnetic systems offer elegant ways to manipulate the essential characteristics of SKs and as such, are playgrounds where the role of topological defects can be best studied. We propose to use this opportunity to explore the consequences of well-controlled perturbations on the global properties of potential SK-host compounds *and* aim for the discovery of new types of magnetic SKs. To that end, we will focus on a selection of systems, where the above contributions can be conveniently discriminated. Under the guidance of theoretical and numerical tools, we will apply reciprocal space methods – namely quantum beam diffraction and spectroscopy– to unravel the important structural and dynamic features of SKs, over extended length and time scales. A peculiar attention will be given on the spin dynamics, which is able to deliver prime information concerning the nature and stability of the studied magnetic structures, as wall as the interaction of conventional excitations (spin waves, phonons) with SKs.

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[10] A. Chacon *et al.*, Phys. Rev. Lett. **115**, 267202 (2015) ; Y. Nii *et al.*, Nature Comm. **6**, 8539 (2015)

[11] See A.A. Tereshchenko *et al.*, Phys. Rev. B **97**, 184303 (2018) and references therein.

Experimental and numerical techniques – Our research program will make a massive use of quantum beam methods and macroscopic characterization techniques. While the former reveal intimate features of matter at the atomic scale, the latter evaluate their impact on the bulk electromagnetic properties. Neutron scattering will be our workhorse to address the main structural properties and dynamic features (in the ps-ns range) of the studied systems. X-ray scattering and spectroscopy will yield access to the materials properties under high pressure. µSR will offer sensitivity to the long-time (≈ µs) dynamics of the ground state magnetic structures and SK lattices. The above techniques are available at large scale facilities located at the "Plateau de Saclay" (LLB, SOLEIL) and in Europe (ILL/Grenoble, MLZ/Munich, PSI/Zurich, *etc.*) in general. In addition, “laboratory” experiments (magnetic and transport measurements, X-ray diffraction and Mössbauer spectroscopy) will permit accurate pre-characterizations *and* grant access to well-targeted features. In combination with the above techniques, we will use theoretical and numerical (Monte-Carlo, DFT, *etc.*) methods to put our experimental results in a broad perspective. This will be done through ongoing collaborations with experts in the field.

Task 1 – Doping-induced SKs and magneto-elastic anomalies in chiral helimagnets. MnGe stabilizes spin helices with very short wavelengths (λ ≈ 30 Å). As we have recently shown, λ can be increased by a factor ≈ 20 upon a 50 % substitution of Mn- by Co-/Rh-ions. In parallel, *doping* triggers the building of arrays of magnetic defects, akin to SK-anti SK pairs, even in zero applied magnetic field[12]. In order to make further progress on that matter, we want to sequentially reach full Mn substitution, which leads to the Pauli paramagnet CoGe and unconventional superconductor RhGe, respectively. By following the micro- and macroscopic properties of this whole series, we will be able to track the evolution of ground state magnetism and the response of magnetic field-induced SKs to quenched *disorder*. MnGe is also characterized by spin-state transitions and strong *magneto-elastic* anomalies, resembling the invar-alloys properties, as revealed by our recent high pressure X-ray experiments[13]. It will be interesting to extend these studies to doped systems and check the survival of these uncanny pressure-induced features.

Task 2 – Topological spin textures in frustrated alloys. Reentrant spin glasses (RSGs) are disordered alloys with competing interactions of general formula $A_{1-x}B_x$. Chemical substitution introduces antiferromagnetic (AFM) B-B interactions within a mostly ferromagnetic (FM) A-A matrix, yielding *frustration*. The B-B pair distribution is controlled by stoichiometry and/or thermal quench. In a recent study of a Ni_xMn_{1-x} alloy[14], we have shown using neutron scattering and Monte Carlo simulations that strongly disordered magnetic defects with fractional charge -akin to SK-anti SK bound states- are stabilized around the B-B pairs under an applied magnetic field. Their size varies as a function of field following well-defined scaling laws and their collapse occur only under very large magnetic fields (of the order of the AFM coupling), which can be as high as several tens of T. We propose to explore the transition from the RSG towards the canonical spin glass (*i.e.* magnetically disordered) regime and evaluate

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[12] N. Martin *et al.*, Phys. Rev. B **96**, 020413(R) (2017)

[13] M. Deutsch *et al.*, Phys. Rev. B **89**, 180407(R) (2014); N. Martin *et al.*, Phys. Rev. B **93**, 214404 (2016)

[14] I. Mirebeau, N. Martin *et al.*, *Spin Textures induced by Quenched Disorder in a Reentrant Spin Glass : Vortices versus Frustrated Skyrmions*, submitted to Phys. Rev. B (2018)

the role and stability of the magnetic defects in this process. This will be done through a study of the series of amorphous FeMn alloys in a large composition range.

Task 3 – Hunt for SKs in triangular magnets. We plan to study the field-induced properties of two magnets, namely $CsCuCl_3$ and $BaCo_2(AsO_4)_2$[15]. These compounds indeed possess the essential characteristics required for the appearance of SKs (namely a strong uniaxial anisotropy and a triangular crystal structure) but have never been studied in this light up to now. This task is prospective but will potentially culminate in the observation of SKs in an entirely new class of systems (see **Sec. 1**).

3. Originality and feasibility of the project. While current research on magnetic SKs is mostly based on real space imaging in thin films or multilayers, our project uses reciprocal space methods to probe properties of the studied samples over their full volume (not only at their surface) *and* under extreme conditions (high pressures, large magnetic field, *etc.*). Combining these experimental techniques with theoretical tools will grant access to the material-specific properties required to improve our control over SKs in condensed-matter *and* lead to a better understanding of the role of topological defects in phase transitions of matter. The principal investigator and his collaborators are fully proficient to the tools required to achieve the proposed scientific tasks. This places our research program on solid grounds and ensures its feasibility.

4. Expected results. The research activity focusing on magnetic SKs is currently exploding, as demonstrated by the abundance of high-level scientific publications (Nature, Science, PRL, *etc.*) and dedicated conferences (especially in Germany, Japan and USA). Based on our recent experience and preliminary results, we target many publications in highly ranked journals, presentations in international events and dissemination to a large audience (summer schools, seminars and outreach).

5. Expected collaborations. Scientific partnership with internationally recognized experts is a key aspect of our project. The proposed work indeed assumes a *strong* and already *well-developed* collaboration network, with a common publication record.

Field	Collaborations
Neutron scattering & spectroscopy	• "Novel Frontiers in Quantum Materials" group (LLB) • "Magnetism and Neutron Diffraction" group (CEA Grenoble) • M.T. Fernandez-Diaz & T. Hansen (Institut Laue Langevin) • L.J. Bannenberg & C. Pappas (TU Delft, The Netherlands) • E. Altynbaev & S. Grigoriev (Petersburg Nuclear Physics Institute (PNPI), Gatchina, Russia)
High pressure X-ray scattering & spectroscopy	• F. Baudelet, J.-P. Itié & L. Nataf (Synchrotron SOLEIL) • M. Deutsch (Université de Lorraine, Laboratoire CRM2)
Muon spin relaxation	• A. Amato & D. Andreica (Paul Scherrer Institut, Switzerland) • F. Bert (Laboratoire de Physique du Solide, Orsay)
Theory & modelling	• U.K. Rössler (IFW Dresden, Germany) • A.O. Leonov (Hiroshima University, Japan) • S. Mankovsky (LMU, Munich, Germany) • P. Bonfà & R. De Renzi (Università di Parma, Italy)
Solid-state chemistry	• C. Decorse (ICCMO, Université Paris-Sud, Orsay) • A. Tsvyashchenko (Vereshchagin Institute, Moscow, Russia)

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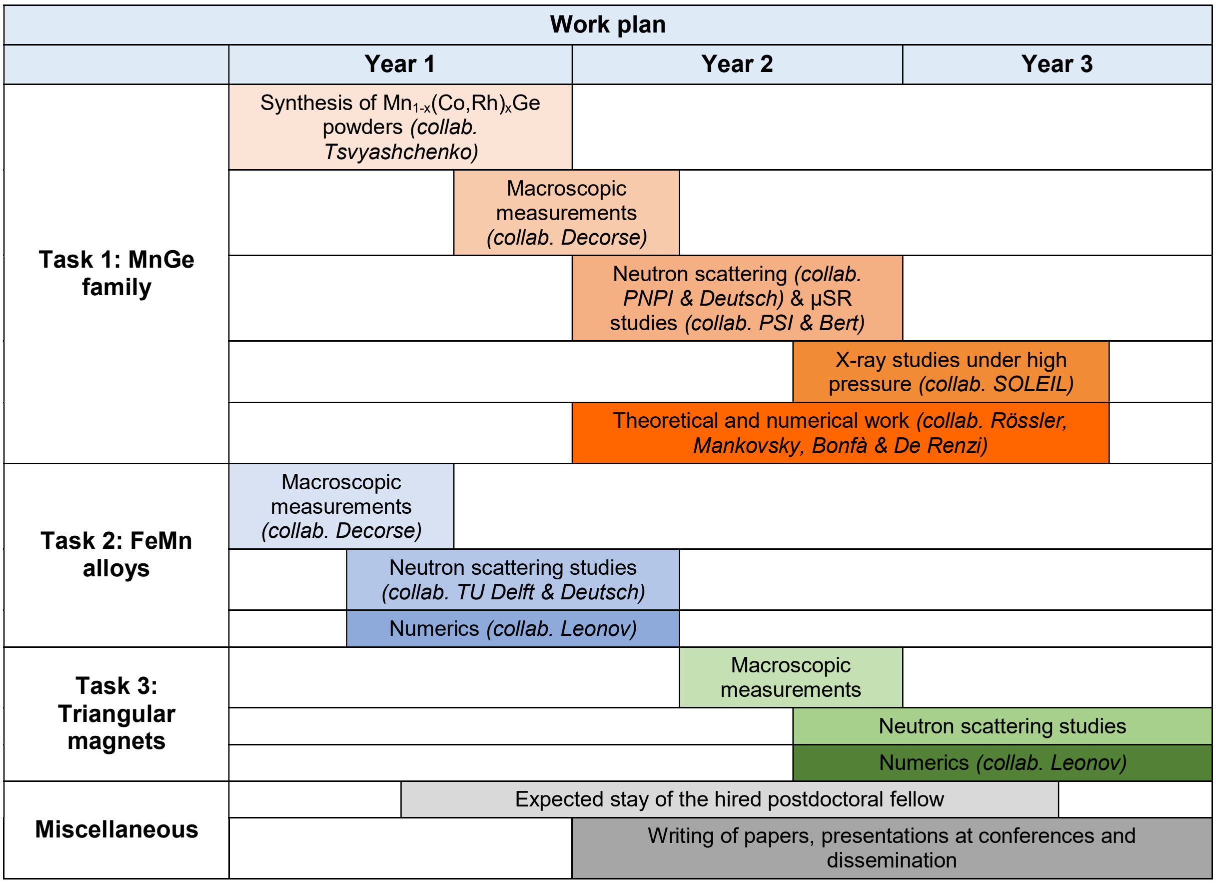
[15] K. Adachi *et al.*, J. Phys. Soc. Japan **49**, 545 (1980); L.-P. Regnault *et al.*, Heliyon **4**, e00507 (2018)

Besides a full-time implication of the principal investigator, the recruitment of a *post-doctoral fellow*, competent in quantum beam techniques (neutrons, X-rays and/or muons), is required. The successful candidate will be involved in all parts of the research work (experiments, data treatment, paper writing and presentation in international events). We believe that the proposed project will be highly beneficial for her/him, namely thanks to the practice of a large array of experimental techniques *and* the participation in a very dynamic research area.

6. Work and financial plan for a 3 years period



Work plan

	Year 1	Year 2	Year 3
Task 1: MnGe family	Synthesis of $Mn_{1-x}(Co,Rh)_xGe$ powders *(collab. Tsvyashchenko)*		
	Macroscopic measurements *(collab. Decorse)*		
		Neutron scattering *(collab. PNPI & Deutsch)* & μSR studies *(collab. PSI & Bert)*	
		X-ray studies under high pressure *(collab. SOLEIL)*	
		Theoretical and numerical work *(collab. Rössler, Mankovsky, Bonfà & De Renzi)*	
Task 2: FeMn alloys	Macroscopic measurements *(collab. Decorse)*		
	Neutron scattering studies *(collab. TU Delft & Deutsch)*		
	Numerics *(collab. Leonov)*		
Task 3: Triangular magnets		Macroscopic measurements	
		Neutron scattering studies	
		Numerics *(collab. Leonov)*	
Miscellaneous	Expected stay of the hired postdoctoral fellow		
		Writing of papers, presentations at conferences and dissemination	

Financial plan

Budget line	Estimated cost*	Comment(s)
Chemical products	**2 k€**	Synthesis of $Mn_{1-x}(Co,Rh)_xGe$ alloys
Access to quantum beam facilities (neutrons, X-rays and muons)	2 x 10 x 1 k€ = **20 k€**	Travel and stay expenses, typically 10 experiments outside of LLB during years 2 & 3
Progress meetings	2 x 5 x 1 k€ = **10 k€**	Visits to collaborators for focused discussions
Software licences	2 x 3 k€ = **6 k€**	For data treatment and simulations
Open-access fees	**8 k€**	High-impact journals like Nature Comm., PRX, *etc.*
International conferences	2 x 4 x 1.5 k€ = **12 k€**	Typically 4 events, in Europe, Japan and/or USA
Online communication	**1 k€**	Design of a website exposing significant results & popularization
Organization of a "end of program" workshop	**20 k€**	Focused on recent theories and experimental results on magnetic skyrmions in bulk materials
Total	**79 k€**	

*Costs which are multiplied by 2 account for the principal investigator and the hired postdoctoral fellow.