Title: Highly Frustrated Magnetism

Acronym: HFM

Principal Applicants

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Abstract

This project is a joint effort between solid-state chemists, experimental and theoretical physicists to unveil novel quantum states and effects where frustration plays a leading role. The main goal is to reach a broad understanding of the important physical parameters that drive these new ground states and sketch out the generic phase diagrams for a broad variety of degrees of freedom. These degrees of freedom extend beyond the simple frustration of magnetic interactions to include lattice couplings, orbital degrees of freedom, dilution effects, electronic doping, etc. Our project represents a timely effort of having a broad general view in a field which can be approached through various systems: highly frustrated antiferromagnetic lattices, orbital liquids, metallic spinels..., where original properties such as spin liquid, spin ice, orbital order and so on...-see below- have been recently discovered. Studying doping in these systems also provides new tools to examine the problem of High T_c cuprates through a combination of the methods that describe a propagating hole in the resonating valence bond states, methods that will certainly provide new concepts in the field of condensed matter.

Keywords

Magnetism, spin liquids, orbital liquids, frustration

Status of the field and research context

Since the discovery of High T_c Superconductivity, the idea of stabilizing new quantum states in correlated systems has been investigated both from the theoretical and the experimental directions. Frustration of antiferromagnetic interactions has been singled out as being a dominant ingredient in this quest for novel states, as proposed initially by Anderson in 1973 in his "resonating valence bond" model (RVB) of the triangular lattice. In the past decade, frustration in lattices that are less coordinated than the triangular one has been shown to lead to a possible stabilization of new states, including the RVB state. Such magnetic networks are observed in kagomé and pyrochlore systems (lattices of corner sharing triangles and tetrahedra respectively) and beyond. In fact stabilization of such exotic and intriguing ground states as spin liquids, orbital liquids, spin ice systems has been found to occur when there is a competition between various degrees of freedom (magnetic, charge, orbital, elastic…) which turns out to lead to unusual effects when lattice frustration is present. Frustration is also of major importance in the physics of correlated fermions. High T_c superconductors were one example of this. The recently discovered superconducting and magnetic cobaltites present another system of growing importance, since frustration of the magnetic interactions is inherent to the triangular structure of the Co network.
We sketch below an overview of the various research fields that are sorted into 4 principal classes: frustration of magnetic interactions which is indeed the most classical and intense axis of study, the lifting of degeneracy of the ground state in frustrated systems and two topics where magnetic degrees of freedom interplay either with orbital or itinerant character. One short section is devoted to experimental approaches: the ability to use a large variety of techniques allows to progress in a concerted way. Theoretical approaches have been developed in close connection with experiments; they have benefited in the recent past from new tools related to advances in computing. Exact diagonalization techniques for S=1/2 quantum spins and Monte-Carlo simulations on classical systems have given major insights into the ground state of these systems. One is now able to include defects (e.g. in kagomé systems), anisotropy and dipolar interactions (e.g. in pyrochlores), which starts to fill the gap between experimental systems and theoretical models.

1. ***Frustration of magnetic interactions***

Among the systems recently studied by our groups from both the experimental and theoretical directions, some have already given rise to collaborations at the European level. In more details, one can cite:

*Kagomé lattice antiferromagnet*s:
Because of their simple 2- dimensional archetypal corner sharing geometry, they have been extensively studied analytically and numerically over the past ten years using either classical approaches or exact diagonalization techniques (in the case of spin S=1/2). They represent a simple playground for the RVB scenario and a very original singlet ground state has been predicted with an unusual continuum of singlet excitations filling the gap to the triplet state. The presence of such a continuum of excitations might be at the origin of a power law behavior of the specific heat, insensitive to any applied field, wheras the susceptibility which reflects the population of the triplet state should be gapped. The extensive number of low lying states which can be retrieved by classical approaches, features a liquid-like behavior and, indeed, the highly fluctuating character and anomalous specific heat of the ground state, first detected in the corner-sharing S=3/2 compound, $SrCr_9Ga_3O_{19}$ (SCGO), triggered a major experimental interest in this kagomé bilayer material, and variants discovered subsequently. Magnetic susceptibility, correlations and excitations have been studied in a refined manner. They are certainly the closest to the realization of the pure Heisenberg hamiltonian on a corner sharing lattice among the geometrically frustrated magnets. Despite an impressive amount of data, the central search for a well defined gap has not yet concluded although the susceptibility measured by NMR decreases at low T in SCGO. Also, the existence of an original spin glass-like fluctuating ground state, not related to the defects of the structure, is now evidenced. The explanation of this state is now a central open issue for more elaborate theories accounting for the dynamics in the ground state.

Other systems have been investigated in detail, such as jarosites, where spins lie in simple kagomé planes rather than in kagomé bilayers. In these materials couplings that have yet to be understood typically cause transitions towards conventional magnetic states at low T. An exception appears to be hydronium jarosite where particular single-ion anisotropy appears to play a key role in the stabilisation of an unconventional spin glass phase.
Definitely, the most challenging avenue today is the search for a pure S=1/2 kagomé compound where the number of defects, whatever their nature, are minimized. This is a central issue for material scientists and a first step towards a textbook comparison to theorist predictions has been achieved through the rediscovery of volborthite, a natural mineral which presents a kagomé geometry made of isosceles triangles. This compound is synthesized and being deeply investigated by several groups of the proposed collaboration .

Pyrochlore lattice antiferromagnets
They are the 3D analogues of the 2D kagomé lattice materials, in which the network of corner-sharing tetrahedra provides an appropriate geometry for achieving a spin liquid state. The Heisenberg pyrochlore antiferromagnet is the 1st example of 3D spin liquid; for quantum spins it is expected to behave like the kagomé system, with a spin gap and a continuum of low energy singlet states. Such a geometry is much more common than the kagomé one: it can be found in pyrochlore type compounds ($A_2B_2O_7$) and in spinel systems $A_2B_2O_4$; it is also found in the rare earths-intermetallics systems with Laves Phase structure, RT_2. Many compounds of these series have been synthetized and studied in recent years, with both rare earth and transition metal ions at the magnetic sites. This is still a broad field to be explored since the use of many elements of the periodic classification enable the investigation of the effects of many parameters, such as the value of the magnetic moment (values between J=1/2 and J=15/2 can be studied) or the effect of anisotropy, which can be planar or axial. Magnetic ordering is often observed, due to additional interactions (see section 2: lifting of degeneracy), but several compounds do not order at low temperature and could be 3-dimensional spin liquid systems.

J_1/J_2 models on square lattices
Frustration can also be caused by a competition between 1st neighbour interactions, J_1 and 2nd neighbour ones, J_2. Although in these systems theory is far more advanced than experiments, recent experimental realizations in vanadates $Li_2VO(Si,Ge)O_4$ have enabled the first tests of the theoretical predictions for this model. These vanadates are characterized by a ratio J_2/J_1 of the order of unity and recent experiments have demonstrated the possibility to vary the degree of frustration by applying hydrostatic pressures.
The relevance of the spin-lattice coupling in relieving the ground-state degeneracy, the effects of spin dilution and the possible stabilization of superconductivity by charge doping are some of the studies envisaged on these systems. Also, from the theoretical point of view, several aspects of the J_2-J_1 phase diagram need to be clarified. For example, it is not yet established how the nature of the ground-state changes on approaching the critical value $J_2/J_1=0.5$, or how the phase diagram is modified upon increasing the spin value. Moreover, the evaluation of the finite temperature thermodynamic properties for J_2/J_1 around 0.5 is a serious theoretical challenge, since certain powerful numerical approaches as Quantum Monte Carlo, cannot be used.

Cu Delafossites
They are layered oxides with a stacking of triangular-based, S=1/2 planes. Although they have long been studied in the classical stoichiometry $R^{3+}B^{+}O_2$, the major recent opening in the field of magnetism came from the possibility of inserting additional oxygen anions into $R^{3+}(=La^{3+},Y^{3+})Cu^{+}O_2$ compounds established in 1994 by Cava et al.. Recent developments in the synthesis of these materials by the Grenoble group have eliminated stacking faults and enabled the first experimental studies of what might prove to be the first hole-doped highly frustrated antiferromagnetic system.
In undoped systems, $YCuO_{2.5}$, $YCuO_{2.66}$ and $LaCuO_{2.66}$, various geometries of Cu^{2+}, S=1/2 magnetic lattices can be realized, including both one dimensional for the former and planar arrangements of corner sharing triangles for the two latter systems. Doping can be achieved by simple heterovalent Y^{3+}/Ca^{2+} substitutions and, indeed magnetic properties are found to change with such doping. The ground states obtained in this new family can vary from dynamical to antiferromagnetic order and a better understanding will come from a theoretical and experimental study of the couplings in this system.

2- Lifting of degeneracy

In these strongly degenerate systems any "small interaction" is able to remove the degeneracy and must consequently be accounted for. The latest developments in this field lie in the terms that perturb the pure Heisenberg Hamiltonian such as magneto-elastic coupling, dipolar interactions, anisotropy and Dzyaloshinsky-Moriya couplings. These terms remove the ground state degeneracy and lead to a wealth of novel ground states including spin ice, topological spin glass, cooperative paramagnetism, etc. This competition between the alternative ground states and possibility that allows a system to retain its residual entropy have given rise to tremendous theoretical activity in this field for classical systems which presently dominates the more traditional classical and quantum approaches developed for the purely Heisenberg case.

Particularly important is the coupling to lattice degrees of freedom, as is commonly observed for example in the insulating spinels. Here, phonons can gain energy from the presence of frustrated bonds, leading to novel magnetoelastic transitions; the coupling of the phonons to the huge number of low-energy degrees of freedom of the magnetic system can then give rise to unusual dynamical phenomena of the hybrid magnetoelastic degrees of freedom. As always, the many instabilities inherent in frustrated magnetism can lead to rich phase diagrams when further external parameters such as pressure or magnetic field are varied.

3- Magnetization plateaux

Frustration can also have spectacular consequences on the magnetization curve of antiferromagnets: It can induce magnetization plateaux at rational values of the magnetization. This has been predicted and observed, for instance, by members of our collaboration in spin ice $Dy_2Ti_2O_7$ and observed in $SrCu_2(BO_3)_2$, a realization of the 2D orthogonal dimer model, for which plateaux have been observed at 1/8, 1/4 and 1/3 of the saturation value. Plateaux have also been predicted in other frustrated models, like the kagome lattice, both at T=0 for spin 1/2 and at finite temperature for classical spins. These plateaux are expected to correspond to a broken symmetry phase. This was confirmed by an NMR investigation of the 1/8 plateau of $SrCu_2(BO_3)_2$, which has revealed a large number of different Cu sites, hence a large supercell. In systems consisting of coupled dimers, like the orthogonal dimer model, triplet excitations on dimers behave as a gas of hard-core bosons, the magnetic field plays the role of the chemical potential, and the transition into a plateau is similar to a metal-insulator transition. In that respect, frustrated magnets constitute a new playground to study the properties of strongly correlated hard-core bosons. The investigation of the consequences of this analogy is just beginning. Very recently, observation of a transient magnetization plateau was observed in a S= 1/2 Cu Kagome lattice; this could be related to a different mecanism: a dynamical order by disorder mecanism. It is clear that in frustrated systems the proximity in energy of many different phases could be at the origin of such plateaux.

4- *Orbital liquids*

Frustration induced by interactions between orbital degrees of freedom can lead either to orbital order or to orbital liquid states. In transition metal compounds with (almost) degenerate 3d orbitals spin and orbital degrees of freedom occur with equal measure, and superexchange or double exchange interactions involve both of them. Charge carriers in such systems can couple to both spin and orbital excitations which makes the physics of such systems as CMR manganites and other doped transition metal oxides very rich. In (undoped) Mott insulators orbital interactions influence strongly the spin interactions, and can induce frustration even in non-frustrated lattices, for instance on a 3D cubic lattice, leading to interesting new behavior and to competition between different ordered and disordered (quantum liquid) states. Examples of such behavior, currently investigated both in experiment and in theory, are $LiNiO_2$, $LaTiO_3$, $BaVS_3$, or some spinel compounds. Furthermore, frustration of various multipolar interactions is not confined to d-electron systems: Rare earth and actinide compounds show a great variety of strongly fluctuating states, including the novel heavy fermion state of rare-earth-filled skutterudites, which arise from the unresolved competition of different kinds of order.

5- *Metallic spinels and metallic pyrochlores*

In metallic spinels and pyrochlores, frustration influences not only the spin degrees of freedom, but also those associated with charge. Various behaviors have been observed, which can be influenced by frustration: metal-insulator transitions (e.g. in $Tl_2Ru_2O_7$), superconductivity ($Cd_2Re_2O_7$), or anomalous

Hall effect ($Nd_2Mo_2O_7$). It was also proposed that heavy-fermion like behavior can be induced by frustration: the prototype compound is LiV_2O_4 with mixed valence V ions; there is now need for more compounds and physical studies in order to understand the physics of these systems which could be at the border between the spin liquid and Fermi liquid states.

6- *A panel of experimental physical methods from our collaboration*

The magnetic complexity of these systems necessitates a wide array of experimental approaches of different character and of complementary nature. To reveal the competition between ground states, one requires well-characterized samples. The chemical routes will be developed in our objectives part. Regarding physical studies, access to large-scale facilities dedicated to neutron scattering, muon spin resonance, for which the number of proposals in the field of frustrated magnetism have increased considerably in the past few years, and synchrotron radiation are important for such studies and this is an area where the European community has a dominant lead over North America and Japan.

Sub-kelvin cryogenics and standard techniques of macroscopic nature as specific heat, magnetization and transport measurements are widely employed to characterize the basic properties of the investigated systems.

Beyond these, many groups of our collaboration are specialized in quite advanced techniques either of local/resonant nature or covering the reciprocal space.

Local probe advanced techniques such as μSR and NMR brought major breakthroughs in the recent past. Implanted muons allow to probe the spin dynamics at low T (~10 mK) in any system. Both the small coupling of the muon to its magnetic environment and the time-window make it a unique probe of liquid states and quantum T-independent relaxation processes in kagomé and fluctuating pyrochlore systems. In a quite complementary manner, due to strong coupling with the surrounding magnetic ions, nuclei are invaluable for probing local susceptibility and local magnetic structures with a high sensitivity. NMR appears as a forefront tool to disentangle the role of defects of the magnetic lattice such as spin vacancies or bond defects, and the intrinsic behavior of the susceptibility associated with geometric frustration such as in SCGO. NMR, as well, brought major breakthroughs in the study of J_1-J_2 vanadates such as the characterization of the magnetic ground state, a detailed study of the spin susceptibility and, thanks to the sensitivity of the hyperfine coupling to the local structure, the observation of frustration driven lattice distortions. The access to the High Field Grenoble facility enabled one group of the collaboration to reveal the structure of the magnetization on the 1/8 plateau in $SrCu_2(BO_3)_2$ at 28 T and 35 mK.

Neutrons are an essential probe to study Highly Frustrated Magnetism at the microscopic level. They provide a direct access to the magnetic correlations both at the short range scale (inter atomic distances) through the diffuse scattering, and at the long range scale (100nm or greater) through the Bragg diffraction. For instance, members of our collaboration successfully used neutron scattering to investigate spin-ice behavior and pressure induced magnetic order microscopically. Moreover, inelastic neutron scattering allows one to measure the low energy spin excitations (typically in the 10 μeV to 10 meV range), which are a key characteristics of these systems and allow direct comparison with theoretical calculations of the magnetic response function. Both dispersive excitations in magnetically ordered systems and cluster-like excitations in spin-liquid or spin-glass systems can be characterized in detail over a wide range of energy and length scales..

Raman scattering has a very high sensitivity for phonon excitation and sufficiently low energy resolution for magnetic scattering required to distinguish systems with broken-symmetry ground states from liquid-like states. One can determine lattice distortions and elucidate the relevant energy scales of magnetic/electronic excitations. Systematic Raman scattering investigations of a large number of compounds and doping studies have been performed in groups within our collaboration. Further, collective or exciton-like magnetic bound states with singlet character that exist in Highly Frustrated Magnets can be observed in Raman scattering. The derived information about the singlet dynamics is complementary to the triplet spectrum investigated using neutron scattering. The possibility of a direct observation of collective orbital excitation (orbitons) is still strongly debated. Here, more experimental and theoretical effort is presently invested to increase our understanding.

Objectives and expected benefit from European collaboration

The previous experimental and theoretical state of the art sketches the broad diversity of highly frustrated magnetism which encompass many of the novel electronic states discovered in the recent years. Of course, our daily route is to synthesize, study experimentally and refine models with systems as perfect as possible and where physical relevant parameters can easily be singled out. Beyond this, the ambitious goal of our network is to sketch some generic phase diagrams of frustrated systems where lattice and orbital degrees of freedom are taken into account and spin vacancies and/or itinerant holes bring new physical controlled parameters. This is still a long way in the future and our major collaboration axis requires theory, material synthesis and physics experiments to be closely coordinated. The recent developments in the expanding field of highly frustrated magnetism, as signalled by the success of the conference "Highly Frustrated Magnetism" held in Grenoble last August, makes this gathering of efforts quite timely. We expect that European collaboration will accelerate interdisciplinary research in contrast to the current situation where individual countries can only enter the field slowly. It is worth noticing that many sporadic collaborations are already established most frequently between two individual groups, here and there, never more. Our network will provide a great stimulation and synergy to our research and will allow us to compete, at the european level with our american and japanese colleagues. Specialized workshops are often organized in Japan, and in the US there is at least one topical meeting per year at the APS; thus ESF support would allow European researchers to reach the same intellectual stimulation. In addition, this program will help to integrate scientists from the new member states of the European Union; finally an important component of the program is related to education and training of students and post-docs.
A typical outcome of such a broad collaboration between experimentalists is the possibility to work on samples which are characterized by several techniques. This allows to reach an unambiguous interpretation of the experimental results and leads to a deeper understanding of the physical properties underlying these systems.

Below, we sketch some to-date open problems, emerging new theoretical concepts and synthesis approaches which are obviously at the heart of our objectives and future progresses in the field. This short list should be considered as some initial guidance for future European scale work but we indeed expect from our meetings to offer novel routes and allow to bend the initial goals of our collaboration in a constantly moving field.

<u>Open problems</u>

We list here various key-questions regarding the topics mentioned in the state of the art section
- Kagomé antiferromagnets: the transition temperature to the low T spin-glass behavior is not related in a simple manner to the concentration of spin vacancies. Is there an intrinsic spin glass behavior in disorder free geometrically frustrated magnets?
- J_2/J_1 model on a square lattice: it is a real challenge to synthesize or vary physical parameters so as to get near the critical $J_2/J_1 = 0.5$ value. Liquid state is expected for such a ratio of interactions. The study of the dilution and electron doping effects is also envisaged.
- Pyrochlores: only at very low T, order is present in $Tb_2Ti_2O_7$. Is this an ideal spin liquid and why does it behave so? Estimation of interactions (exchange, dipolar, anisotropy) would lead to a magnetic ground state. Interesting as well is the comparison with $Tb_2Sn_2O_7$. What is the influence of the metallic and non magnetic ion?
- Delafossites: the unexpected magnetic transition in $RCuO_{2.66}$ requires a better understanding of the couplings and appropriate models to be built on reliable grounds.
- Orbital problems: in spinel systems, orbital degrees of freedom play a big role and are coupled with lattice distortions. Further studies are necessary to clarify the interplay between orbitals, spins and distortions.
- Impact of frustration in metallic systems, including the fashionable cobaltites. Many questions have to be answered: Is an RVB state description the most appropriate? Is superconductivity favored by frustration? What is the influence of metallicity on the spin liquid state?
- Magnetization plateaux: They have been predicted for several lattices, including the kagome lattice, but observed so far only in some specific geometries. The access to High Manetic Field European

facilities is certainly central for this search. From the theoretical point of view, the physics outside the plateau remains quite mysterious and is difficult to understand within the present theories.

New topics and concepts, applications

Fractional spin phases

Quantum effects are ubiquitous in magnetic materials, yet they remain poorly understood. Fractionalization of the excitations is one of them. In Europe, a few teams have already done some noticeable work on this subject (both experimentally and theoretically) which remains widely open. In a fractionalized system, the quasiparticles carry unusual quantum numbers; for example, instead of a magnon with spin S=1, one finds pairs of unconfined spinons with spin S=1/2. Such electron fractionalisation goes along with exotic quantum spin liquid phases. These systems escape any kind of simple ordering transition all the way down to zero temperature, but they may exhibit some unusual properties related to topological exotic order, involving macroscopically ranged quantum correlations which should resist to quantum decoherence. Frustrated magnets are ideal candidates for exhibiting such liquid phases as their geometry often precludes conventional magnetic ordering. Extensive study of the inelastic spectrum of these RVB compounds would be an interesting step to increase our understanding of these RVB phases. The thermodynamic properties of the triangular S=1/2 compound Cs_2CuCl_4, which have recently been studied in Japan and in Marseille, let suppose that they might be good candidates for spin liquids and would deserve more microscopic studies: inelastic neutrons, X ray scattering, muon spectroscopy, NMR....

More generally, the study of excitation spectrum of various systems will help to classify and to distinguish the various types of possible behaviors.

Magneto-caloric effects and applications

The largely unexplored area of finite field effects in frustrated magnets possesses not only a great fundamental interest but also has a high potential for new technological applications. A dramatic change of entropy during a transformation from a non-degenerate fully polarized state at high fields to an infinitely degenerate state at low fields leads to an enhanced magnetocaloric effect. Adiabatic demagnetisation of a frustrated magnet will, therefore, produce a large decrease in its temperature or magnetic cooling. An enhanced magnetocaloric effect suggests that frustrated spin systems are prospective refrigerant materials and can some day rival at low temperature conventional paramagnetic salts for use in adiabatic demagnetisation refrigerators, which are widely considered for space applications and promise to develop into a new generation of environmentally friendly refrigerators.

Synthesis: novel and more traditional routes

From a solid state chemist point of view there is no obvious possibility to design solid structures in a predictive and systematic way and this is of course also true for low-dimensional quantum spin systems. One has therefore to rely on some few concepts, like for example dimensional reduction or naïve pictures that have emerged based on experiences with structurally related materials.

A concept that has proved to be promising for finding new compounds with a low dimensional arrangement of magnetically interesting ions is to synthesize oxides and oxohalogenides containing so called lone-pair ions *e.g.* Pb^{2+}, Sb^{3+}, As^{3+}, Se^{4+}, Te^{4+}. Such ions have a stereochemically active lone-pair and thus a more or less one-sided coordination. The lone-pairs act as scissors to cut down the dimensionality of compounds when they are present. An example is $Cu_2Te_2O_5Br_2$ which has weakly connected Cu^{2+} tetrahedra.

An approach based on the use of the inherent structural flexibility of particular frameworks has been quite successful in the case of vanadates. Indeed, within the transition metal oxides systems, vanadates exhibit a unique structural chemistry due to its capacity to realize different vanadium-oxygen coordination schemes associated with different oxidation states 3+, 4+ and 5+. In addition, it is possible to isolate a very large number of mixed-valence compounds. In the frame of quantum spin system, the number of vanadium based compounds (V^{4+}, S = ½) has increased tremendously now to overtake the cuprates. From the structural point of view it is possible to obtain phases formed of isolated polyhedron (in the form of vanadium 4+ square pyramids) like in $K_2(VO)V_2O_7$, dimers formed of edge connected square pyramid pointing up and down as observed in $VOSeO_3$, isolated chains like in $MgVO_3$, 2D layers as in CaV_4O_9 or more exotic structure as for example for $Na_2V_3O_7$. In addition to this structural versatility, vanadium oxides present also a bench of unusual electronic properties with complex behaviour that are based on the interplay of charge, spin, orbital and lattices degrees of freedom. The idea is to extend this approach to other systems with particular spins arrangements (Cu^{2+}, Ti^{3+}, Ni^{2+}). Any phases composed of appropriately corner-connected, essentially rigid polyhedral units could potentially show such inherent structural flexibility.

Minerals are also a great source of inspiration for the synthesis of new low dimensional or frustrated materials. Examples are: azurite $Cu_3(OH)_2(CO_3)_2$, a one-dimensional diamond chain; malachite $Cu_2(OH)_2CO_3$, a S = ½ quantum antiferromagnet with a spin gap; stibivanite Sb_2VO5, a S = ½ 1D chain; dioptase $Cu_6Si_6O_3.6H_2O$, a S = ½ dimer system. Traditional oxide-based systems including variants (doped, substituted or non-stoichiometric) of pyrochlores, spinels, garnets, and delafossites (*e.g.* $Er_2(Ti/Sn)O_7$, $Gd_2Ti_2O_7$, $ZnCr_2O_4$, $ZnCr_2Se_4$, $Gd_3Ga_5O_{12}$ $CuFeO_2$). Also compounds showing structures with Kagomé like arrangements are of great interest *e.g.* $Cs_2B_{12}Cu_3F_{12}$; $K_3Cu_3P_2$, Jarosite $KFe_3(SO_4)_2(OH)_6$, and volborthite $Cu_3V_2O_7(OH)_2.2H_2O$.

Within the network we have access to a wide variety of preparation methods including solid state synthesis, crystal growth from melts and growth from vapour phase. The main techniques are: gas phase transport reactions, hydrothermal synthesis, flux techniques, high pressure techniques, floating zone with infrared image furnace. Special techniques for growing single crystals from melts are Bridgman, top seeded solution growth, flux and Czochralski methods. Many special furnaces allowing controlled gas pressures, extreme pressures etc are at our disposal. The sample preparation is accompanied by detailed thermodynamic and thermoanalytic investigations, chemical analysis, microstructure and crystal structure determination.

European context

To our knowledge, there are currently no network programs on a European level having financial support on the topic of highly frustrated magnetism. The complexity of this topics requires close collaboration between scientists of different fields, including chemists, physicists, theoreticians.

Several international collaborations between 2 or 3 groups involved in our proposal have been very successful in the past years and we intend to extend such collaborations to a larger scale, so as to reach the critical size requested for a high efficiency.

The access to large scale facilities (neutrons, μSR, high magnetic fields) is essential for our proposal and a coordination at the european level will ensure the highest quality of the proposals in the field of HFM.

Finally, we would like to point out that in this field European teams have often been the leaders (spin ice behavior, $CsCu_2Cl_4$, SCGO were first discovered in Europe) and the collaboration between the participants will ensure that European teams can compete efficiently with USA and Japan in this rapidly progressing field.

Work plan

Program management:

Program management will proceed from tight contact between the partners through meetings, e-mail, seminars and a Web-site which will allow an enlarged diffusion of information in the field of Highly Frustrated Magnetism. We hope that new groups, starting to work in this field will join our program during the 5 years.

Scientific Activity

As stated above, scientific activity relies upon the present state of the art. The forefront activity which is crucial to the field now is the synthesis of novel materials which will approach better the ideal quantum liquid spin state. Combining sample characterizations and a wide array of advanced techniques such as NMR, μSR and neutron scattering will enable detailed studies of the new systems for which theoretical modelling is an absolute necessity in order to make new advances. Attention will be paid also to cross-disciplinary applications of these studies as, for instance, in regards of the protein folding processes.

Exchange of researchers

Exchange of researchers is important when electronic communication reaches its limits, *ie*, brainstorming about new systems, analysis and discussion of the data. It is especially important to us that PhD students can also travel to various collaborative groups in order to achieve a more complete scientific expertise and to participate in various experimental work.

Workshops
Two kinds of meetings will be organized:
- small workshops (about 20 participants) on a well defined topics (2 days). Planning of these workshops will depend on advances in our activities in the field and does not require much administrative work. Every year 2 topical meetings could be organized.
- Large workshops involving all groups (about 60 to 80 participants) with both invited and contributed talks (3 days). The steering committee will be held on these opportunities. Two large meetings could be organized during the 5 years of the project: one midterm, and a final meeting.

We also plan to organize a school.

Duration

5 years

Budget estimate (per year)

Exchange visits and complementary support of travel expenses to large scale facilities : 40 000 €
Science meetings: 30 000 €
Advertising and Website: 5000 €
External administrative costs: 5000 €
Total per year: 80 000 €
Besides the regular activities, a school will be organized for 40 to 50 participants: 90 000 €

Full coordinates and CV of the applicants

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1992-1993: Postdoc at the department of Physics, Hiroshima (Japan)
1993-2000: research associate, Phys. Institut, RWTH Aachen

2000: Habilitation at RWTH Aachen
2000-2002 Deputyship Prof. in Physics, RWTH Aachen and Univ. of Technology Braunschweig
From 2003: Research associate at MPI for Solid State Research
Number of papers: 87 + 6 review articles or chapters in a book, 36 invited talks
Research topics: Magnetism and strongly correlated electron systems, low dimensional and frustrated spin systems, optical spectroscopy, ultrasonic spectroscopy

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Curriculum vitae
1986-1989: JRF at Lincoln college , Oxford University
1989-1994: position at ILL, Grenoble
From 1994: University College London. Professor of physical Chemistry, from 2000
Research publications: 58 research papers, 2 comments, 9 reviews, 3 conference abstracts
Research topics: Solid State chemistry and condensed matter physics: experiments (particularly neutron scattering) and theory (analytical and numerical); discovery of "spin ice" in 1997

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1988: Doctor of the Physical Sciences
Since 1968: employed by the Solid state department of Central Research Institute (Budapest)
1972 – 1974: post-doc at the Cavendish laboratory (Cambridge)
1980 – 1984 and 1988-1991: senior research fellow at University of Cologne
1991- 19993: Senior research fellow at ICTP (Tieste)
1998: full professor at the Budapest University of Technology and Economics
Research topics: theory of strongly correlated electrons systems, heavy fermions, triangular antiferromagnets, quantum spin liquids, multipolar order in skutterudites and actinide compounds

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1993: PhD at the Department of Structural Chemistry, Stockholm University
1994-2001: Project leader at the Department of Inorganic Chemistry (Stockholm university)
From 2001: Research associate in inorganic chemistry, Stockholm University
Publications: 49 Journal articles , 1 book Chapter, 10 patents, 15 refereed conference proceedings, 18 conference abstracts.
Research experience: synthesis of ceramic whiskers, ceramic composites. Synthesis, structural characterisation and magnetic properties of low-dimensional compounds.

List of the 5 most relevant publications of the applicants during last 5 years

- J.M.D. Champion, M.J. Harris, P.C.W. Holdsworth, A.S. Wills, G. Balakrishnan, S.T. Bramwell, E. Cizmar, T. Fenell, J.S. Gardner, J. Lago, D.F. McMorrow, M. Orendac, A. Orendacova, D.M. Paul, R.I. Smith, M.F. Telling, A. Wildes; $Er_2Ti_2O_7$: evidence of order by disorder in a frustatred antiferromagnet; Phys. Rev. B 68, 020401 (2003)
- K. Penc, M. Mambrini, P. Fazekas, F. Mila: Quantum phase transition in the SU(4) Spin Orbital model on the triangular lattice, Phys. Rev. B 68, 012408 (2003)

- P. Lemmens, K.-Y. Choi, E. E. Kaul, Ch. Geibel, K. Becker, W. Brenig, R. Valenti, C. Gros, M. Johnsson, P. Millet and F. Mila,: Evidence for an unconventional magnetic instability in the spin-cluster compound $Cu_2Te_2O_5Br_2$, Phys. Rev. Lett. 87, 227201 (2001)
- P. Carretta, R. Melzi, N. Papinutto and P. Millet: Very low frequency excitations in frustrated two-dimensional s=1/2 Heisenberg antiferromagnets, Phys. Rev. Lett. 88, 047601 (2002)
- P. Mendels, A.Keren L. Limot, M. Mekata, G. Collin and M. Horvatic: Ga NMR study of the local susceptibility in $SrCr_8Ga_4O_{19}$: pseudogap and paramagnetic defects, Phys. Rev. Lett., 85, 3496 (2000)
- A. Keren, Y.J. Uemura, G. Luke, P. Mendels, M. Mekata and T. Asano: Magetic dilution in the geomatrically frustrated $SrCr_{9p}Ga_{12-9p}O_{19}$ and the role of local dynamics: a μSR study, Phys. Rev. Lett. 84, 3450 (2000)
- L. Limot, P. Mendels, G. Collin, C. Mondelli, B. Ouladdiaf, H. Mutka, N. Blanchard, M. Mekata: Susceptibility and dilution efefcts of the kagomé bilayer geometrically frustrated network: a Ga NMR study of $SrCr_{9p}Ga_{12-9p}O_{19}$, Phys. Rev B, 65, 144447 (2002)
- P. Carretta. N. Papinutto, C.B. Azzoni, M.C. Mozzati, E. Pavarini, S. Gonthier, and P. Millet "Frustation driven structural distortion in $VOMoO_4$" Phys. Rev. B 66, 094420 (2002)
- P. Lemmens, M. Grove, M. Fischer, G. Güntherodt, V.N. Kotov, H. Kageyama, K. Onizuka, Y. Ueda, Collective Singlet Excitations and Evolution of Raman Spectral Weights in the 2D Spin Dimer Compound $SrCu_2(BO_3)_2$, Phys. Rev. Lett. 85, 2605 (2000).
- S. T. Bramwell and M. J. P.Gingras,Spin Ice State in Frustrated Magnetic Pyrochlore Materials, *Science* **294** 1495 (2001).
- K. Kodama, M. Takigawa, M. Horvatic, C. Berthier, H. Kageyama, Y. Ueda, S. Miyahara, F. Becca, F. Mila Magnetic superstructure in the Two-Dimensional Quantum Antiferromagnet $SrCu_2(BO_3)_2$. *Science* **298**, 395 (2002).
- F. Becca and F. Mila , Peierls-Like Transition Induced by Frustration in a Two-Dimensional Antiferromagnet.*Phys. Rev. Lett.* **89**, 037204 (2002).
- S. Dommange, M. Mambrini, B. Normand, F. Mila , Static impurities in the kagome lattice: dimer freezing and mutual repulsion., Phys. Rev. B **68**, 224416 (2003).
- G. Ehlers, A. L. Cornelius, M. Orendac, M. Kajnakova, T. Fennell, S. T. Bramwell, J. S. Gardner, Dynamical Crossover in 'Hot' Spin Ice, *Journal of Physics - Condensed Matter*, **15**, L9 - L15 (2003).
- E.Ya. Sherman, P. Lemmens, B. Busse, A. Oosawa, H. Tanaka, Ultrasonic attenuation on the Bose-Einstein transition in $TlCuCl_3$, Phys. Rev. Lett. **91**, 057201 (2003).
- P. Lemmens, K.Y. Choi, G. Caimi, L. Degiorgi, N.N. Kovaleva, A. Seidel, F.C. Chou, Giant phonon anomalies in the pseudo-gap phase of TiOCl, cond-mat/0307502, subm. to Phys. Rev. Lett., (2003).
- I. Kézsmárki, Sz.Csonka, H. Berger, L. Forró, P. Fazekas, and G. Mihály : *Pressure dependence of the spin gap in* $BaVS_3$, Phys. Rev. B **63**, 081106(1-4) (2001).
- A. Kiss and P. Fazekas, *Octupolar ordering of* Γ_8 *ions in magnetic field*, Phys. Rev. B **68**, 174425 (2003).

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