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Recent Trends in Magnetism and Superconductivity

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ABSTRACT BOOK

Invited lectures

Spin-dependent hybridization and spin polarization effects at metal-organic interfaces: Ferrocene- and pyrene-based cyclophane chemisorbed on Co(111) nanoislands

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I will discuss an approach to molecular spintronics based on aromatic molecules chemisorbed on ferromagnetic surfaces. The chemisorption is governed by strong hybridization of molecular π-orbitals with *d*-states of the substrate. The spin-split band structure of the latter leads to different hybridization for spin-up and spin-down states. This spin-dependent hybridization induces a spin-imbalanced density of states in the molecule, resulting in an induced magnetic moment for strong or spin-filter properties for weak coupling [1]. The hybridization also modifies the moment, exchange interaction, and anisotropy of the involved substrate atoms, which eventually can lead to magnetic hardening [2], *i.e.* the molecule and the substrate atoms directly bound to it form a hybrid molecular magnet with enhanced coercivity and blocking temperature [1]. We investigated the potential utilization of polycyclic aromatic hydrocarbons containing cyclophanes as building blocks for single-molecule hybrid spintronic devices. For this purpose, we designed and synthesized a novel cyclophane based on pyrene (Py) and ferrocene (Fc) [Fc-Py^2-Fc, inset in Fig. (a)] that exhibits a unique ecliptic aromatic plane arrangement with an interplanar distance of 3.52 Å. In the framework of spin-dependent hybridization the lower aromatic plane of such a structure is expected to form a hybrid molecular magnet, while the upper level retains sharp, but spin -polarized by hybridization, electronic states that can generate spin filter functionality [1,2]. We have succeeded for the first time in sublimating such a cyclophane on ferromagnetic double-layered Co nanoislands on Cu(111) and studying it at the single-molecule level by spin-polarized scanning tunneling microscopy (SP-STM) [3]. The comparison of topographic STM images of Fc-Py^2-Fc [Fig. (a)] and a likewise synthesized reference molecule [Fc-Py-Fc, Fig. (b)] on both Cu substrate and Co nanoislands suggests strong chemisorption of the lower Py with the Co surface atoms accompanied by a significantly reduced conductance through the Py-stack. SP-STM measurements of Fc-Py^2-Fc on ferromagnetic Co nanoislands [Fig. (c)] did not indicate any spin-filter effect induced by the Py-Py stacking. This can be explained by chemisorption-induced renormalization and spatial downshift of the lower π-system, both of which lead to weaker π-π coupling [3]. Intriguingly, an almost equally strong but reversed spin polarization was observed over the Fc units compared to the pristine Co surface, suggesting that Fc-based wires could be promising spin polarizers.

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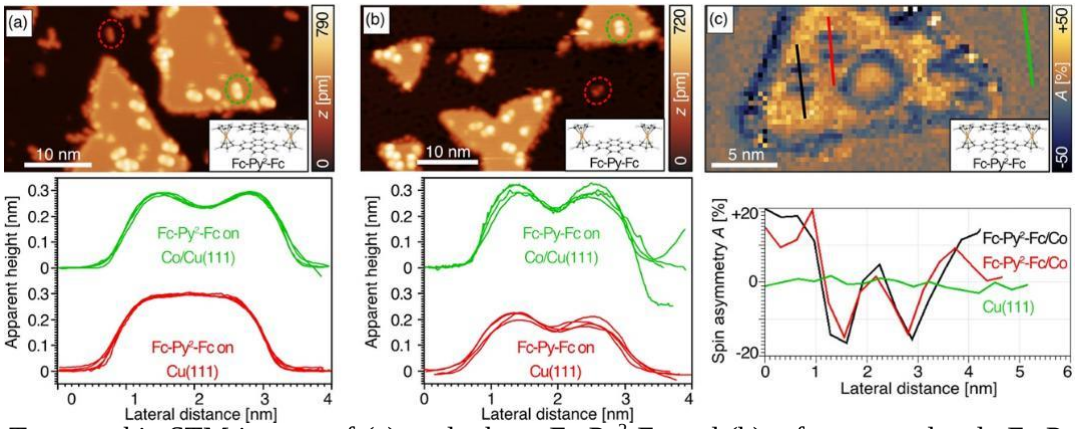

Figure: Topographic STM images of (a) cyclophane Fc-Pc^2-Fc and (b) reference molecule Fc-Py-Fc on Cu(111) and Co/Cu(111). (c) Spin asymmetry map $A(x,y)$ of cyclophane Fc-Pc^2-Fc on Co/Cu(111). The lower panels show profiles along the longitudinal molecular axes of the molecules in (a,b) or along the lines in (c), both marked with the corresponding colors in the STM images.

Exploring nanoscale magnetism in ferrimagnetic spintronic materials, with soft x-ray spectroscopy, scattering, and imaging techniques

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Ferrimagnetic materials offer a highly versatile and tunable platform for spintronics research. In particular, rare earths (RE)- transition metals (TM) amorphous thin films alloys consist of antiparallelly coupled magnetic sublattices which exhibit perpendicular anisotropy, a magnetic compensation temperature and, eventually, a spin reorientation transition. Through the choice of elemental constituents, stoichiometry and thickness variations, their magnetic properties can be controlled, enabling functionalities for particular application in magnetic storage and sensorics[1,2].
In the first part of the talk I will discuss the anomalous 'wing shape' hysteresis loop which occurs close to the compensation temperature of ferrimagnetic thin films. It refers to a counter-intuitive effect that manifest itself as an apparent decrease of the net magnetization that takes place when the magnetic field overcomes a certain threshold. The shape of the side magnetic loop bears the characteristics of an intrinsic exchange bias effect. Analysing the elemental magnetic moments for the surface and the bulk parts, separately, permits to identify a temperature window where this effect takes place. It also allows to discriminate among previously proposed models, supporting a vertical domain wall formation as a response of the system towards an energetically favourable ground state[3].
In the second part of the talk I will dwell on magnetic skyrmions. Magnetic skyrmions are stabilized by the Dzyaloshinskii–Moriya interaction (DMI), originating from the inversion symmetry breaking, as for instance in centrosymmetrical B20 crystals. They exhibit fascinating magnetic properties and dynamics within a narrow temperature pocket. For applications, ferrimagnetic skyrmions offers advantages, as for instance the possibility to control the skyrmion Hall angle in a robust magnetic environment. To this end, we report on the observation and characterisation of ferrimagnetic skyrmions and their domain walls in DyCo films. These skyrmions, with antiparallel aligned Dy and Co magnetic moments have a characteristic core radius of about 40 nm, and are formed during the re-magnetization process. They were identified by combining X-ray magnetic scattering, scanning transmission X-ray microscopy, and Hall transport technique[4]. Moreover, through XMLD imaging the domain walls are shown to be largely of Néel type, supporting he hypothesis of an intrinsic DMI may occur in the bulk part of ferrimagnetic amorphous layer.
Finally, I will briefly present preliminary results involving simultaneous measurements of current induced magnetic switching and element specific magnetic spectroscopy on ferrimagnetic Hall bars. In a spin-orbit torque (SOT) switching experiment, a charge current is passed through a heavy metal (Pt) with a large spin Hall angle that further generates a pure spin current in the lateral direction. This spin current is injected in the adjacent magnetic material. Above a threshold current density, the SOT can electrically switch the FiM/AFM sublattice magnetization. However, high current densities may not only switch the magnetic order but through anisotropic thermal gradients and voltages may also lead to an apparent re-magnetisation process. Therefore, a direct measure of the magnetic evolution through the XMCD and/or XMLD spectroscopy as a function of the write current is presented as a new method for spintronics[5].

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Ultrafast optical generation of magnetic texture in antiferromagnets

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Recent experimental demonstration of skyrmion nucleation with terahertz lase [1] has opened new horizon in ultrafast manipulation of long-range magnetic order. Its theoretical understanding is, however, still under the mist. So far, the theoretical studies in this direction are limited to magnetisation classical magnetisation dynamics, which completely overlooks the fast magnetisation dynamics. We overcome this hurdle by employing a hybrid quantum-classical evolution scheme [2,3] which consider both electronic and magnetic degrees of freedom on equal footing. Our approach is capable of looking into the fast electronic dynamics within first 100fs as well as explore the slow magnetisation dynamics taking place after several picoseconds. We successfully identified the emergent interaction responsible for the long-range order and also estimate the life time of the emergent configurations. We further combine a dynamic topological characterisation to study the evolution of the topological features. Our result shows that the emergent topological texture appears in pairs with equal and opposite topological invariants conserving the total topological charge.

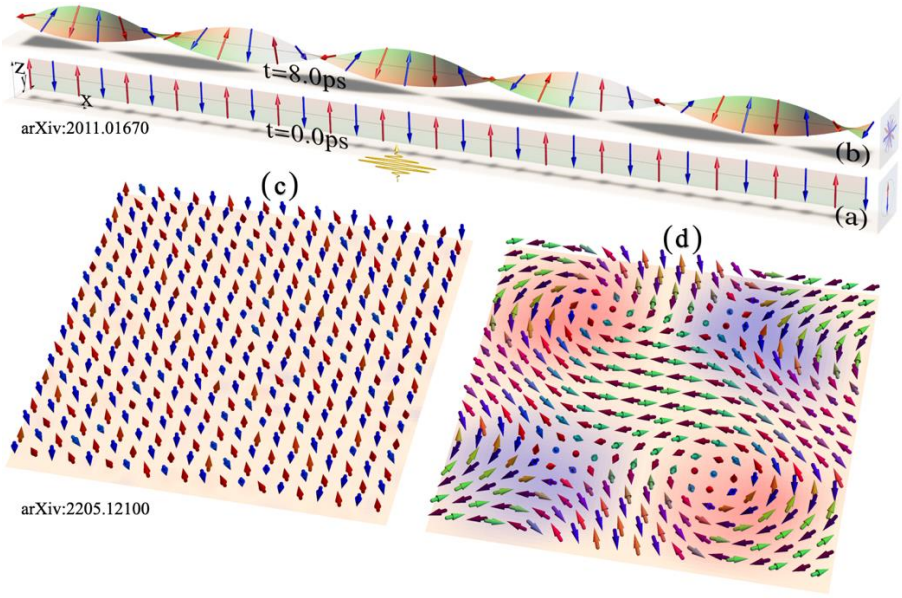

Fig. Formation of antiferromagnetic (a,b) spin spiral and (c,d) meron-antimeron pair with ultrafast laser.

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Memristive effects in multiferroic metal-organic heterostructures

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Versatile multi-level memristive elements based on multiferroic tunnel junctions have interesting features determined by the possibility to modulate the junction's electrical resistance by independently controlling the magnetization orientation of the ferromagnetic electrodes (the tunneling magnetoresistance) and the ferroelectric polarization of the tunnel barrier (the tunneling electroresistance) [1]. The coupling of these magneto-electric properties is one of the most active field of research in materials sciences opening a large spectrum of technological applications from nonvolative memory, to elements in logic circuits, sensing devices, energy harvesting, biological synapses models in the emerging area of neuromorphic computing and artificial intelligence. Realizing these multifunctional electronic elements using organic materials and/or biomolecules is presenting various advantages related to their low cost, versatile synthesis, flexibility, lightweight and biocompatibility that are actually of major scientific and technological interest for applications in the fields of molecular electronics, bioelectronics, and therapeutic techniques. Moreover, the lightweight elements in their composition showing a small spin-orbit coupling, favor to maintain for longer times than in standard inorganic materials the electron spin state, *i.e.* the spin lifetime of the charge carriers, combining advantages for the applications in spintronics and molecular electronics.
Herein, we demonstrate the memristive spintronic behavior of multiferroic tunnel junctions in elements with two terminals composed of nanoscale tunneling barrier consisting on organic biomolecular ferroelectric films of guanine nucleobabses [2] sandwiched between two different ferromagnetic electrodes of Co and Co_xCr_{1-x} films with in-plane easy axes of magnetization. The realization of these memristive states is controlled by the characteristic properties of the ferroelectric and the magnetic films. Typical ferroelectric polarization–electric-field hysteresis loops showing large electrical polarization and pyroelectric signal of guanine are observed at low temperatures up to 200 K. Above this transition temperature, at which different properties are affected, the guanine films have a preponderant paraelectric phase containing residual or locally induced nanoscopic ferroelectric domains, as observed by piezoresponse force microscopy at room temperature.

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Scanning tunneling spectroscopy of quantum critical and topological magnets: Searching for the link between electronic band structure and magnetism

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Scanning tunneling microscopy and spectroscopy is a powerful tool to study the electronic density of states. Many studies in superconductors show the opening of the superconducting gap in the density of states. This has been very helpful to characterize relevant parameters such as gap magnitude and shape of the density of states in many systems. However, it remains very difficult to associate features in the density of states to bulk magnetism. Here I will discuss millilkelvin scanning tunneling spectroscopy experiments in three magnetic systems, Rh doped CeRu2Si2, Co3Sn2S2 and EuCd2As2. Ce(Ru0.92Rh0.08)2Si2 is a heavy fermion which presents an antiferromagnetic phase (AFM) below 5 K. At very low temperatures, AFM vanishes above 2.5 T, and one finds a correlated paramagnetic phase. When further increasing the magnetic field, the latter becomes, above 5.5 T, a field induced polarized magnetic phase. I will discuss the influence of Kondo hybridization and Zeeman splitting on the magnetic phase diagram. Furthermore, I will discuss the evolution with magnetic field of the flat band in Co3Sn2S2 and the topological properties of EuCd2As2 in the ferromagnetic phase.

Trends and opportunities in two-dimensional (2D) materials spintronics

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Spintronics, exploiting the spin degree of electrons as the information carrier, is an active research field for implementing the beyond-CMOS devices.
Recently, two-dimensional (2D) materials have been attracting enormous attention in spintronics owing to their unique spin-dependent properties, such as the ultra-long spin relaxation time of graphene [1] and the spin–valley locking of transition metal dichalcogenides. Moreover, the 2D materials-based heterostructures provide an unprecedented possibility of combining different characteristics (spin-orbit coupling, superconductivity, semiconductor band-gap engineering, ferromagnetism, etc) via the proximity effects [2, 3].
This talk will overview some of the most recent progress in spintronic research focusing on 2D materials and related heterostructures to systematically summarize the progress of spin injection, transport, manipulation, and application for information storage and processing. The talk will also highlight the current challenges and future perspectives on the studies of spintronic devices based on 2D materials.

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Novel broadband and efficient THz radiation sources based on spintronic structures

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The field of THz spintronics is a novel direction in the research field of nanomagnetism and spintronics that combines magnetism with optical physics and ultrafast photonics [1]. The experimental scheme of the field involves the use of femtosecond laser pulses to trigger THz emission by exciting ultrafast spin and charge current dynamics in thin multilayers. The films are composed of ferromagnetic (FM) and non-magnetic (NM) thin layers where the non-magnetic layer features a strong spin-orbit coupling. The technological and scientific key-challenges of THz spintronic emitters are to increase their intensity and to shape the frequency bandwidth. To achieve this the control of the source of the radiation, namely the transport of the ultrafast spin- and charge current is required.
In this presentation, I will address the generation, detection, efficiency and the future perspectives of THz emitters. I will present the state-of-the-art of efficient emission in terms of materials, geometrical stack, interface quality and patterning. The different factors that define the strength of the THz emission like the spin Hall angle, spin diffusion length, conductivity and interface transmission will be quantified and correlated with combined THz and GHz spectroscopic measurements.

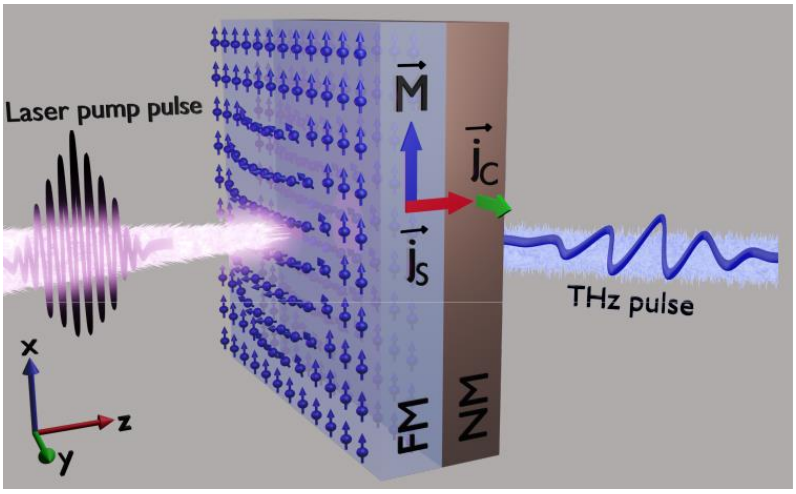

Figure 1: The field of THz spintronics involves the use of femtosecond laser pulses to trigger THz emission by exciting ultrafast spin and charge current dynamics in thin ferromagnetic/non-magnetic multilayers.

The impressive so far results render spintronic THz emitters a desirable new class of THz sources possessing promising advantages like versatility, durability, ease of operation, high pump-power scalability, and broadband THz emission with tunable polarity.

E. Papaioannou, R. Beigang, Nanophotonics 2021

Superconducting and hybrid passive shields for magnetic field mitigation

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Superconductors have demonstrated unique properties as low-frequency magnetic-shielding materials. They can indeed provide a very-low field background for ultra-sensitive devices or a strong magnetic mitigation over a short distance, cutting down electromagnetic compatibility problems. Both active (i.e., coils fed with appropriate currents [1]) and passive layouts (i.e., simple superconducting cavities [2]) were successfully employed, the latter ones consisting of superconducting (SC) bulks and/or SC coated conductors/tapes [3,4]. Among SC materials, MgB_2 is a promising option because the precursors are cheap and non-toxic, the low weight density makes it suitable for space applications and the long coherent length allows the fabrication of large untextured polycrystalline samples with high critical current densities.
In this work, we investigated the shielding ability of MgB_2 bulks shaped as hollow-cylinder with a capped end (henceforth called cups) with an aspect-ratio of height to diameter approaching one, meeting the practical requirement of great shielding performance in small dimension shields. These shields were manufactured by Spark Plasma Sintering of MgB_2 powders added with hexagonal boron nitride powders, an approach allowing the fabrication of bulks fully machinable with cutting tools [5].
The experimental measurements of the shielding factors (SFs) carried out in the axial-field orientation evidenced a remarkable screening ability with SFs exceeding 10^4 and 10^2 near the closed extremity and in the inner half of the cup, respectively, up to a field threshold of 1.8 T at the temperature T = 20 K [6]. However, these shields are affected by two weaknesses: the strong decrease of the shielding capability when the field is not applied parallel to the main axis of the layout (due to the small aspect-ratio) and the occurrence of thermomagnetic instabilities. To overcome these limitations, both these features were investigated using a 3D numerical model based on a vector potential (**A**)-formulation [7]. The numerical analysis highlighted that the addition of a ferromagnetic shell is very efficient in improving the SFs of the superconducting shield when the applied field is tilted with respect to the shield axis. Moreover, this effect can be maximized if a vertical mismatch of the open end of the two cups is considered [8]. In such a way, a SF rise from 20 to more than 100 is predicted in the whole inner half of the shield. On the other hand, the coupling of the above mentioned A-formulation based model with a thermal model was proved to be very efficient in predicting the flux-jump phenomena, guiding us in the optimization process of the shield and of its thermalization.

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Twisted van der Waals Heterostructure SQUIDs Fabricated by Dry Transfer of 2D Superconductor Flakes

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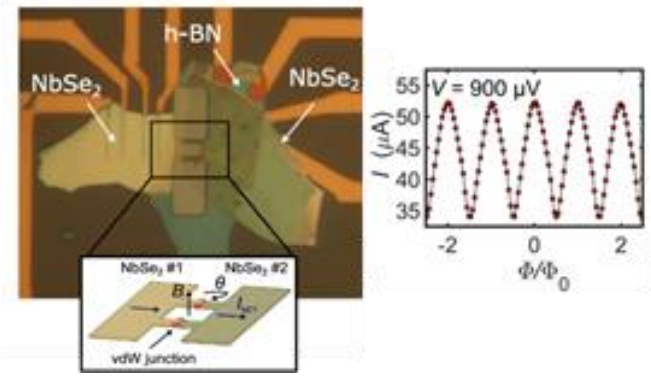
It is well known that a dissipationless supercurrent can flow across a dielectric tunnelling barrier between two superconducting electrodes due to the Josephson effect. Here we describe recent progress in forming Josephson junctions and SQUIDs by exploiting coherent transport across the *twisted* interface between two 2H-$NbSe_2$ flakes that are in intimate contact [1, 2]. A dry transfer technique has been developed to exfoliate, stack and deterministically misalign the flakes in a high purity glovebox environment. Structures are encapsulated with thin flakes of hBN to allow removal from the glovebox for further device processing and measurement without degradation in air. We find that the Josephson dynamics of the resulting twisted $NbSe_2$-$NbSe_2$ junctions is very sensitive to the misalignment angle of the crystallographic axes, allowing control of the hysteresis in I-V characteristics. A single lithographic step has been used to shape junctions into SQUID geometries with typical loop areas of ~25 μm^2. These devices display strong critical current (I_c) oscillations as a function of applied magnetic field with large stable current and voltage modulation depths. The single crystalline structure of these SQUID elements, along with their low defect densities, may offer circuit components with superior performance, e.g., as qubits. Very similar fabrication protocols can be used to make other types of high performance devices and this will be illustrated by our recent work to develop encapsulated graphene Hall sensors for scanning probe microscopy in ambient environments.



(Left) SQUID fabricated from two $NbSe_2$ flakes at a twist angle of ~25°. (Right) I_c as a function of flux in the SQUID loop at a bias voltage of 900 μV.

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The correlation between the second magnetization peak and the magnetic relaxation rate in superconductors

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The activity of the vortex lattice inside a type-II superconductor is often analyzed by means of the normalized magnetic relaxation rate, also indicated as creep rate (S), which is typically obtained by means of time dependent magnetic measurements. The suppression of the critical current density Jc in a material is associated to an increase of the creep rate and related to the decrease of the efficiency of the pinning centers in blocking the flux quanta movement in the material. Therefore, the understanding of the mechanisms underneath the evolution of the relaxation rate with the temperature (T) and magnetic field (H) can be helpful in improving the electrical transport properties of the superconducting materials. Magnetic relaxation measurements on Iron Based Superconductors (IBS) have been reported to show high values of the creep rate S, together with the presence of a plateau in the S(T) curve, in analogy with the behavior observed in High Temperature Superconductors (HTS). Moreover the region defined as the plateau in the S(T) curve shows a minimum, which appears also in the S(H) behavior. However, these peculiarities in the creep rate have been reported often in concomitance with a Second Magnetization Peak (SMP) in the magnetic hysteresis loop M(H), and therefore with an anomalous modulation of the Jc(H) dependence, independently of the class of superconductor analyzed. In our work we will focus our attention on a correlation detected between the onset of the SMP phenomenon and the minimum in the creep rate, by starting from the magnetic relaxation of a $FeSe_{0.5}Te_{0.5}$ crystal showing a SMP phenomenon, and then extending our analysis to other superconducting materials

Pinning potential in superconductors from multi-harmonic AC susceptibility response

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From economics point of view, larger market penetration of cryo-electro-magnetic devices fabricated from superconducting bulk/wires/tapes depends on the cost, expressed in Euro/kA·m, hence, the price reduction can be achieved by increasing the critical current I_c (maximum current that can be transported without losses).
High magnetic fields strongly reduce critical current density due to the dissipation caused by the movement of flux lines (Lorentz force, thermally-activated flux creep). The above-mentioned dissipation can be minimized by artificial defects (nanostructuring) that could fix (pin) the magnetic flux lines due to the decrease in the free energy if the flux line "sit" on the defect. In the case of $YBa_2Cu_3O_x$ (YBCO) high-temperature superconducting films, nanotechnology of pinning centres involves several approaches: substrate decoration, quasi-multilayers and multilayers, targets with secondary phase nanoinclusions, and combinations of the above mentioned [1], involving various architectures and nanoinclusions. Most of the work worldwide involves the calculation of critical current density and bulk pinning force from DC magnetization loops, or from transport measurements. We introduced a simple, straight-forward method of estimating the frequency-dependent critical current density by using frequency-dependent AC susceptibility measurements, in fixed temperatures and DC magnetic fields, from the positions of the maxima in the dependence of the out-of-phase susceptibility on the amplitude of AC excitation magnetic field. The results are compatible with a model that stipulates a logarithmic dependence of the pinning potential on the probing current. A mathematical derivation allowed us to estimate from the experimental data the pinning potentials in various samples, and in various DC magnetic fields. In the case of several YBCO films with various architecture of artificial pinning, the resulted values indicate large pinning potentials, leading to very small probability of magnetic flux escaping the pinning wells, hence leading to very high critical currents in high magnetic fields [2]. The same approach was used successfully, for the first time to our knowledge, in the case of a $CaKFe_4As_4$ pnictide superconducting single crystal.
Acknowledgements: Work done in collaboration with I. Ivan, A.M. Ionescu, D.N. Crisan (NIMP Bucharest), A. Andrei (Natnl. Inst. Lasers, Plasma and Radiations Physics, Bucharest), A. Galluzzi, M. Polichetti (Univ. Salerno, Italy) and J. Mosqueira (Univ. of Santiago de Compostela, Spain). Financial support from EU POC Project 28 (REBMAT) and COST Actions Nanocohybri and Hi_SCALE is gratefully acknowledged.

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Multifunctional Ferromagnetic Shape Memory Materials: Magnetocaloric, Magnetorezistive and Temperature Memory Effects

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Intensive studies of Ferromagnetic Shape Memory Alloys (FSMA) are justified by the martensitic and also magnetic order-disorder transformations exhibited, leading to a plethora of multifunctionalities of interest for both fundamental science and technological applications.
The martensitic transformation and the Curie temperatures may be tailored by doping the alloys with other elements or by suitable thermal treatments, so that alloys with concomitant or sequential structural and magnetic phase transitions may be obtained. Previous studies showed the possibility of tuning the transformation temperatures and the transition heat for off-stoichiometric Ni-Fe-Ga or Ni-Mn-Ga alloys with or without additions [1 - 5].
In the present work we address the influence of substitutions (Co, Cu, Nd and Al) on the magnetocaloric and magnetoresistive effects in Ni-Fe-Ga prepared as bulk and also as rapidly quenched ribbons and subsequently subjected to different thermal treatments. Also, investigations on the thermal memory effect exhibited by Ni-Fe-Ga FSMA [6, 7] are discussed. The investigation methods used include X-ray diffraction, differential scanning calorimetry, magnetometry and transport measurements. The results highlight the differences between the bulk and the ribbons (as prepared and annealed) and the important role of substitutions.

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The role of mechanically induced defects on the magnetic coupling in metamagnetic shape memory alloys

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Magnetic shape memory alloys (MSMAs) undergo macroscopic deformations upon the application of an appropriate stimulus: temperature, stress and/or external magnetic fields. Since the deformation is rapid and contactless, these materials are being extensively investigated for many technological applications, such as sensors and actuators, energy harvesting and damping devices, among others. These materials also exhibit a giant magnetocaloric effect, whereby they are very promising for magnetic refrigeration. The applications in which they can be used are extremely dependent on the material properties, which are, in turn, greatly conditioned by the structure, atomic ordering and magnetism of the material. Indeed, Ni-Mn based Heusler alloys are MSMAs that have received much attention due to the exceptional properties they show as a result of the coupling between magnetism and crystal structure. In particular, Ni-Mn-Z (Z = In, Sn, Sb) metamagnetic shape memory alloys show a large variation on the magnetization (ΔM) associated with the occurrence of a first order thermoelastic martensitic transformation (MT) between ferromagnetic austenite and weak-magnetic martensite. The large ΔM across the structural transformation makes the induction of the reverse MT possible by the application of a magnetic field [1]. Due to the different magnetic features of the structural phases, such magnetically-induced transformation may give rise to a very large inverse magnetocaloric effect [2] or to giant magnetoresistance [3], thus making these materials very attractive for magnetic refrigeration and sensing applications.

The MT characteristics and the magnetic properties (and therefore the functional properties) of these alloys can be tuned by varying the composition of the alloy, changing the long-range atomic order through thermal treatments or modifying the microstructure of the alloy. Concerning compositional variations, it has been shown that doping the alloy with Co enhances the magnetism of the austenite, thus increasing the ΔM associated to the MT and therefore giving rise to larger magnetically-induced shifts of the MT temperatures and higher associated magnetocaloric effect [1].

On the other hand, it has been shown that the magneto-structural properties of Ni-Mn-In and Ni-Mn-In-Co alloys can be properly tuned varying the long-range atomic order whereas in the Ni-Mn-Sn system the high stability of the L21 structure precludes the modification of atomic order by means of conventional thermal treatments [4].

Despite the aforementioned promising features, the poor mechanical properties Ni-Mn based Heusler alloys exhibit (i. e., brittleness and fragility) hinder their development towards practical applications. As a consequence, the use of microparticles as micro-actuator elements is attracting increasing interest as an effective method in order to surpass the bulk limitations. Moreover, the study of microstructure and microstructural defects has special relevance in the mechanical production of micro and nanoparticles. This is an interesting issue as long as composites consisting of a ductile polymer matrix with MSMA in powder (where the polymer provides integrity and the MSMA the functionality) have been recently proposed and developed to overcome the intrinsic brittleness of the bulk alloys [5]. Likewise, a controlled powder production is also crucial for bed binder jetting or similar 3D printing techniques, which are currently being considered as an alternative for the elaboration of different and complex geometry shape memory alloys -based devices [6]. In this sense, a previous and complete characterization of the powder alloys is needed.

Therefore, in the talk I will present mainly the influence of mechanically-induced defects on the magnetic coupling between the main carriers of magnetic moment in ternary and quaternary metamagnetic Heusler alloys.

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Revised Kittel theory for ferromagnetic domains and further developments

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Kittel's model for ferromagnetic domains [1,2] is still a cornerstone in micromagnetism. It starts with the very simple assumption of a infinite sample extent over two directions and with and a finite thickness d, though much larger than the typical domain size. The domains have the width l over one in-plane direction (i. e. perpendicular to the sample depth coordinate) and infinite extent over the other in-plane coordinate. It is then shown that the surface magnetostatic energy scales with ${M_0}^2 l$ ($\pm M_0$ being the magnetization of the domains) and that the energy corresponding to 180° domain walls scales with $w_{wall} d/l$, where w_{wall} is the wall energy per unit area of the wall and it comprizes the uniaxial magnetic anisotropy constant K_v (which has units of volumic density of energy), the exchange interaction between neighboring atoms J and the interatomic distance a, such that $w_{wall} \propto (JK_v/a)^{1/2}$. By minimizing the magnetostatic + wall energy one derives Kittel's scaling law of the domain size vs. the sample thickness $l \propto d^{1/2}$. There are several aspects that stimulate one to re-visit Kittel's theory of ferromagnetic domains: (i) Kittel's derivations from Refs. [1,2] are rather brief, several aspects (in particular, the sample geometry) are not explicit. (ii) Kittel's derivations are realized in CGS unis, or nowadays everything is expressed in SI units. (iii) In Ref. 1, Kittel computes the average value of the square of the intensity of the magnetic field, $\langle \int H^2 dz \rangle$, while in Ref. 2 the scalar product between the magnetization and the intensity of the magnetic field $\langle - \int \boldsymbol{H} \cdot \boldsymbol{M} dz \rangle$, with the minus sign, is computed. (iv) In Ref. 1 both faces of the sample are taken into account, whereas in Ref. 2 only one face is considered. (v) Kittel didn't write down a complete formula involving the expression of the wall energy in terms of the uniaxial magnetic anisotropy constant and the exchange integral. (vi) The model supposes the existence of a perpendicular magnetocrystalline anisotropy with the easy axis oriented along the normal to the sample's surface, but one needs to take into account also the dipolar interaction energy, which is responsible for the shape anisotropy.

By performing detailed computations, using Kittel's assumption $l \ll d$, one obtains interesting results:

a) The energy from Ref. 1 is in fact half of the energy from Ref. 2, by considering in both cases both faces of the sample; the energy from the evaluation of $\langle - \int \boldsymbol{H} \cdot \boldsymbol{M} dz \rangle$ is in line with Kittel's derivations [2].

b) The energy from Ref. 1 can be brought to the same value as that from Ref. 2 (and in line with Kittel's values) by considering the whole space for the field density of energy, and not only the inner part of the sample;

c) In fact, the magnetostatic volumic density energy has to be written as [3]:

$$w = \frac{\boldsymbol{B} \cdot \boldsymbol{H}}{2} = \frac{B^2}{2\mu_0} - \frac{\boldsymbol{B} \cdot \boldsymbol{M}}{2} = \frac{\mu_0 H^2}{2} + \frac{\mu_0 \boldsymbol{M} \cdot \boldsymbol{H}}{2} \quad (1)$$

This means that one has to consider the energy from Ref. 1 with the energy from Ref. 2 with minus sign, thus in the approximation $l \ll d$ the total magnetostatic energy yields zero value! This finding requires a more detailed derivation of the magnetostatic energy. This can be easily achieved if one considers all solutions for the intensity of the magnetic field inside and outside the sample. The net result (for the volume density of energy, equal to the surface density of energy divided by d) is:

$$w_{tot.} = \frac{8\mu_0 {M_0}^2 l}{\pi^3 d} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^3} \exp\left[-\frac{(2m+1)\pi d}{l}\right] + \frac{w_{\text{wall}}}{l} \quad (2)$$

The minimization of this energy yields the dependence of the domain width with the sample thickness. An important parameter with dimensions of distance comprises the magnetic anisotropy energy, the exchange integral, the spin of one atom S and the interatomic distance a:

$$d_0 = \frac{\pi^2 w_{\text{wall}}}{8\mu_0 {M_0}^2} = \frac{\pi^3 S}{8\mu_0 {M_0}^2} \left(\frac{2JK_v}{a}\right)^{1/2} = \frac{\pi^3 (2JK_v a^{11})^{1/2}}{32 S \mu_0 {\mu_B}^2}$$

(3)

where it was supposed that $M_0 \approx 2S\mu_B a^{-3}$, i. e. one spin S carries a magnetic moment $2S\mu_B$ (μ_B is the Bohr magneton), the gyromagnetic factor is 2 and this magnetic moment corresponds to a volume a^3 of the sample. This parameter has the value of about 0.8 nm for $a \approx 2.5$ Å, $J \approx 10$ meV, $K_v \approx 4 \times 10^5$ erg cm^{-3} ≈ 2 μeV/atom, $S \approx 1$ and about 80 nm for $J \approx 100$ meV, $K_v \approx 2$ meV/atom (and the same values for a and S).

For $d \lesssim d_0$, the minimization of the energy $\partial w_{tot.}/\partial l = 0$ yields the validity of Kittel's scaling law, whereas for $d \gtrsim d_0$ the dependence $l(d)$ is almost linear. The same is obtained when the dipole-dipole interactions are included; just the two regions with $l \propto d^{1/2}$ and $l \approx a + bd$ are much better delimited. On the other hand, experimental data on samples with perpendicular magnetocrystalline anisotropy [4] reported the validity of Kittel's scaling law up to sample thicknesses of hundreds of nanometers or even some micrometers. This means that the d_0 parameter is in the same range, which in turn implies that the magnetic anisotropy energy is in the range of some meV/atom, or some 10^7 Jm^{-3}. This is in line with theoretical previsions, where the magnetic anisotropy energy is in the range of the spin-orbit interaction [5].

In this case, the Stoner-Wohlfarth model fails to describe the hysteresis cycle of most solids. In this model, the coercive field is in the range of $2K_v/M_0$, and this yields tens of Tesla, unacceptably high. The Curie-Weiss mean field theory also fails to explain the coercive field in ferromagnets, its order of magnitude being of $J\eta S/\mu_B$; again, this yields hundreds of Tesla for usual systems. Therefore, the next step was to investigate whether the evolution of the ferromagnetic domain in an external field could give a better result.

Hence, Kittel's model was applied in the case of successive ferromagnetic domains with different sizes, $l(1+\alpha)$ with magnetization $+M_0$ and $l(1-\alpha)$ with magnetization $-M_0$, αM_0 being the net magnetization of the sample. Hysteresis cycles are derived in several hypotheses: (i) stable size of two adjacent domains with opposed magnetization $2l$ and just the relative weight of them varying when the samples exhibits a net magnetization, no dipole-dipole interaction; (ii) stable $2l$, but including the dipole-dipole interaction; (iii) variable $2l$ with the sample net magnetization, no dipole-dipole interaction; (iv) variable $2l$, inclusion of the dipole-dipole interaction. Some models are able to predict non-rectangular hysteresis cycles with shapes quite similar to experimental ones [4,6], but the smallest coercive field is in the range of $\mu_0 M_0/4$ (μ_0 is the vacuum permeability) which is still too elevated with respect to experimental values (it yields some Tesla). Therefore none of the three basic models (Curie-Weiss mean field theory, Stoner-Wohlfarth theory of magnetization rotation in small magnetic nanoparticles, domain wall migration theory) is able to predict the small values observed in the coercive fields of most ferromagnetic materials. There are hopes that a new theory based on band ferromagnetism could offer an explanation for the smallness of coercive fields [7]. This approach assumes a sudden "switch" between spin up and spin down densities of states at small applied magnetic fields, due to the Zeeman interaction, to the difference between spin up and spin down DOSs at the Fermi energy and to the conservation of the total electron density.

In the case of ferroelectrics the Curie-Weiss theory provides good values for the coercive fields [8] and for small sample thickness the domain size is infinite (the single domain state is the most stable). d_0 values in the case of ferroelectrics is below the interatomic distance, but at the same time Kittel's scaling law is approximately valid for sample thicknesses of tens to thousands of nanometers [9]. This implies that the actual theory must be reformulated for ferroelectric domains, by taking into account also the charge accumulation at surfaces and interfaces [8]

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Influence of fast neutron irradiation on the properties of conventional and high temperature superconductors

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Adding defects to the crystal structure of a superconductor is key to achieve high critical current densities. Pinning centers were introduced by an impressive variety of methods. Fast neutron irradiation is a very efficient one allowing to investigate the very same sample before and after the defect addition.
Neutron irradiation was used to sequentially increase the defect density in Nb_3Sn wires and high temperature superconducting (HTS) coated conductors and single crystals. In all cases, the superconducting transition temperature decreases upon irradiation and the critical current first increases, reaches a maximum and finally degrades at high fluences. The beneficial effect is strongest if pinning is weak in the pristine material where the introduced defects trigger a transition from an nearly ordered to a higly disordered vortex lattice.
In materials where pinning is already strong before irradiation, such as REBCO coated conductors, a beneficial effect is still observed up to a certain fluence. At higher defect concentrations, the critical currents are reduced, which is less understood than the initial increase of the critical current at low defect concentration. It will be argued that a suppression of the superfluid density, as predicted by Homes's law, is responsible for the degradation.
Finally, implications for nuclear fusion based on magnetic confinement of the plasma by superconducting magnets will be discussed. First attempts to devise a mitigation strategy against the degradation caused by the unavoidable neutron radiation implemented by thermal annealing will be presented.

Superconducting transport properties in epitaxial $YBa_2Cu_3O_7$ thin films

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In this contribution, an investigation of the transport properties of superconducting transport properties of thin epitaxial $YBa_2Cu_3O_7$ (YBCO) thin films with incorporated artificial pinning centers is presented. In the present context, the artificial pinning centers are oxide nanoparticles ($BaZrO_3$, ZrO_2, and $La_{0.66}Sr_{0.33}MnO_3$). These are either introduced within the matrix of the chemical solution deposited YBCO films, via an in-situ approach or deposited on single crystal substrates prior to the superconducting film growth. The effect of the nanoparticles on the vortex pinning landscape in the YBCO films is evaluated by critical current density, J_c, determination through transport measurements, in different magnetic field and temperature conditions. A special focus is placed on the field angle dependencies of J_c, which were interpreted using the so-called *vortex path model*. In this approach, the angle-dependent J_c characteristics are described using a statistical mechanical model, which considers that the macroscopic state of the system is given by a multiplicity of pinning microstates, which follow a certain distribution function. The vortices follow energetically favorable paths along pinning sites. These paths consist of an integer number of steps having lengths that obey Gaussian or Lorentzian distributions.

Recent developments of MgB2 superconductor at National Institute of Materials Physics

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Work presents the progress in processing and characterization of MgB2 superconductor achieved at National Institute of Materials Physics. Different additives were used to control and enhance vortex pinning. Some additives have been introduced to induce machining properties in MgB2, a hard and brittle compound. Advances in fabrication of textured (001) MgB2 bulks will be introduced. Presentation will discuss specific features in the shape of the pinning force curves with magnetic field.

Vortex dynamics in type II superconductors: long time scales relaxation measurements

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Superconductors are materials with great potential for a wide range of applications and understanding the underlying physics is of upmost importance. Since thermally activated depinning plays an important role in vortex movement, we analyze the corresponding pinning energies by long-time relaxation measurement $m(t)$ in a constant magnetic field. In the range where $\ln(|m|)$ versus $\ln(t)$ is linear, i.e. not too close to the irreversibility line and for a moderate relaxation time window t_w, we can extract the normalized vortex-creep activation energy $U^*=-T\Delta\ln(t)/\Delta\ln(m)$ averaged over t_w. U^* increases with temperature in the collective (elastic) vortex-creep regime (ordered vortex phase) and decreases for plastic (dislocation mediated) creep (disordered vortex phase).
We analyzed various single-crystal specimens (superconducting cuprates and iron based superconductors) of different pinning strengths which exhibit second magnetization peak appearing on the magnetic hysteresis curves [1]. Then, we investigated $Nb_{0.89}Ti_{0.11}$ alloys thermo mechanically processed by intermediate heat treatments at 900 °C in vacuum and cold rolling which show peak effect or second magnetization peak on magnetic hysteresis curves, depending on the thermomechanical treatment to which they were subjected [2]. The last set of samples studied were YBCO thin films with different coatings, focusing on the influence of the coatings on the superconducting properties of YBCO [3-5].

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New superconductor/ferromagnet heterostructure formed by $YBa_2Cu_3O_{7-x}$ and $CaRuO_3$

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Almost all proposed configurations and practical achievements based on superconductor/ferromagnet (S/F) heterostrucutres focus on s-wave superconductors. However, several attempts targeted also high temperature superconductors, most of them using manganite ferromagnets $LaXMnO_3$ (X: Ca or Sr) and $Y_1Ba_2Cu_3O_{7-x}$ (YBCO). Here we propose $CaRuO_3$(CRO) as a new ferromagnetic material that can be used with YBCO for the fabrication of S/F hybrid structures. We show that a ferromagnetic order can be induced by in-plane tensile strain of about 1.7% in a thin layer (~130 nm thickness) of CRO grown by pulsed laser deposition on epitaxial YBCO film. Detailed magnetic and structural investigations will pe presented. The electronic resistance (*R*) of the heterostructure through the superconducting transition will be investigated as a function of magnetic field (*H*) in perpendicular and paralel geometry.

Tunneling Spin-Filtering Effect through Perpendicularly Magnetized Co-Ferrite Films

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The methods to create spin-polarized electric current is an important technical element in the field of spintronics. The tunneling spin-filtering effect, where the electric current tunneling through a ferro(ferri)-magnetic insulator is spin-polarized because of the difference in the tunneling probabilities between up- and down-spin electrons, is among such methods. This effect is historically observed for in-plane magnetized ferro(ferri)magnetic films at both low temperatures [1, 2] and room temperature [3, 4]. To add a variety to the spin-current sources, we have tried to obtain perpendicularly spin-polarized electric current using the spin-filtering effect through perpendicularly magnetized ferro(ferri)magnetic films.
For the realization of this phenomenon, we have to grow thin insulating ferro(ferri)magnetic films with perpendicular magnetic anisotropy on conductive underlayers, which have a role as bottom electrodes. Moreover, the ferro(ferri)magnetic films have to be thin enough and flat enough to guarantee the tunneling conductivity without leakage current.
In a previous study, we have succeeded in growing ferrimagnetic Co-ferrite ($Co_xFe_{3-x}O_{4+\delta}$) (001) films with perpendicular magnetic anisotropy epitaxially on conductive TiN (001) layers [5]. The perpendicular magnetic anisotropy is indcued here by the in-plane tensile strain, but it turned out that the perpendicular magnetic anisotropy degenerates as the Co-ferrite thickness decreases. In the present study, we found that sufficiently thin "insulative" Co-ferrite (I-CFO) (001) films with perpendicular magnetic anisotropy can be grown on "conductive" Co-ferrite (C-CFO) (001) films, and succeeded in observing the evidence of spin-filtering effect through the perpendicularly magnetized ferrimagnetic films.
A typical sample structure is MgO(001) substrate/I-CFO(20 nm)/C-CFO(20 nm)/I-CFO(t = 0.0 ~ 3.0 nm)/MgO(1.5 nm)/Co(1.0 nm)/{Tb(0.45 nm)/Co(0.56 nm)}$_{15}$/Co(2.0 nm). The bottom I-CFO layer is to reinforce the perpendicular magnetic anisotropy of the C-CFO layer, which acts as a bottom electrode and helps the perpendicular magnetic anisotropy of the middle I-CFO layer, which act as a tunneling layer. The MgO layer is to cut off the magnetic interaction between the middle I-CFO layer and the top Co/{Tb/Co}$_{15}$/Co layer, which is a conductive ferrimagnetic layer with perpendicular magnetic anisotropy having a role as a top electrode and a spin detector. The C-CFO and I-CFO can be grown individually by controlling the cation vacancies under different oxidization conditions. The films were fabricated into cylinders with a diameter of 3 ~ 15 μm to measure current-voltage (I-V) curves and magnetoresistance curves. The spin-filtering effect can be confirmed through the magnetoresistance effect accompanied by the switching between parallel and antiparallel magnetization configurations between the middle I-CFO and top Co/{Tb/Co}$_{15}$/Co layers, under perpendicular external fields.
We have succeeded in observing non-linear I-V curves that support the realization of tunneling conductivity. Magnetoresistance effects that support the realization of the spin-filtering effect were also observed. The magnetoresistance ratio becomes larger as the middle I-CFO layer thickness t increases, and the value at 100 K reaches -20% at maximum. This fact well evidences that the magnetoresistance reflects the spin-filtering effect caused by the middle I-CFO layers [6].
This study has been performed in collaboration with Dr. M. A. Tanaka, Mr. M. Furuta, Mr. M. Morishita, Mr. T. Ichikawa, and Mr. D. Mashimo in Mibu's Laboratory at Nagoya Institute of Technology.

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Ultrafast spin transfer in layered magnetic heterostructures

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Controlling magnetic order at high speeds requires the ultrafast manipulation of the spin degree of freedom, a central goal of spintronics. Changing from field-induced manipulation of magnetic order, where speed is limited by spin precession dynamics, to manipulation by ultrashort light pulses could enhance the speed significantly. Magnetic heterostructures consisting of layers with different spin order provide an additional degree of freedom to tune the response of the system to ultrafast laser pulses.
I will present two examples of heterostructures composed of ultrathin epitaxial antiferromagnetic and ferromagnetic layers, measured by time- and element-resolved x-ray magnetic circular dichroism in reflectivity. The ultrafast demagnetization of Co/NiMn ferromagnetic/antiferromagnetic bilayers after excitation by an infrared laser pulse is accelerated by the antiferromagnetic order in the NiMn layer [1]. This is discussed in terms of bidirectional laser-induced superdiffusive spin currents between the ferromagnetic and the antiferromagnetic layers, which persist shortly after the excitation. In Co/Mn multilayers, we observe a transient ferromagnetic alignment of the otherwise antiferromagnetic Mn layers, induced by ultrashort laser pulses. The timescale of the effect is comparable to the duration of the excitation and occurs before the magnetization in Co is quenched [2]. Theoretical calculations identify optically induced intersite spin transfer and an imbalanced population of Mn unoccupied states caused by the Co interface as the cause for this transient ferromagnetic state.
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Machine-learning design of magnets: from molecules to solids

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The process of ending new materials, optimal for a given application, is lengthy, often unpredictable, and has a low throughput. Here I will describe a collection of numerical methods, merging advanced electronic structure theory and machine learning, for the discovery of novel compounds, which demonstrates an unprecedented throughput and discovery speed. This is applied here to magnetism, but it can be used for any materials class and potential application.
Firstly, I will discuss a machine-learning scheme for predicting the Curie temperature of ferromagnets, which uses solely the chemical composition of a compound as feature and experimental data as target[1]. In particular, I will discuss how to develop meaningful feature attributes for magnetism and how these can be informed by experimental and theoretical results.
Then, I will describe how an accurate description of the structure of materials, which is amenable to be used with machine learning, can o er a quantum-chemistry-accurate description of local properties at virtually no computational costs. The method is not just suitable for building energy models[2], namely force elds to used across a broad spectrum of conditions[3], but also for any other local electronic quantity. These models may then be employed to design new materials, as demonstrated here for magnetic molecules with enhanced uniaxial anisotropy[4].
Finally, I will present a novel rotationally invariant representation for generic vector elds. This can be used to generate linear and non-linear machine-learning models, where the total energy depends both on the atomic position and the vector eld direction[5]. The scheme will be put to the test against a hierarchy of simple spin models, demonstrating an impressive ability to extrapolate away from the training region of the data. Application to complex potential energy surfaces, as those extracted from DFT are then envisioned.

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Exploring the Zoo of Layered Quantum Materials

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There has been a recent rush of predictions of a variety of astonishing phenomena and functionalities that can be discovered in layered quantum materials, from topological superconductivity or disipationless conductivity at the boundaries of 2D topological insulators, to emergent particles with fractional charge and statistics or exotic collective behaviours that arise from strong interactions.
A suitable combination of experimental techniques: STM/STS, Helium Atom Scattering and spin-resolved ARPES will be used to characterize the properties of both 3D layered single crystals predicted to be topological superconductors (1T-$PdTe_2$(100) [1]) or antiferromagnetic Topological Insulators ($MnBi_2Te_4$ (001) [2]), 2D epitaxial monolayers proposed to be Quantum Spin Hall or Excitonic Insulators (1T-$MoTe_2$/gr/ Ir(111) [3]) and 1D nanographene polymers with a topological quantum phase transition [4].

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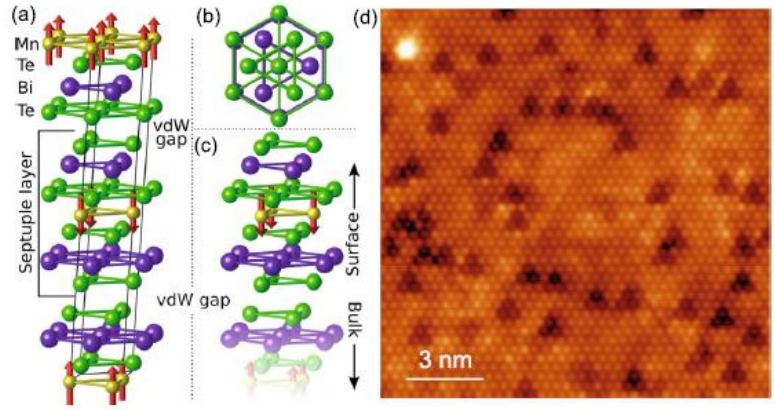

Figure 1: **$MnBi_2Te_4$ bulk and surface structure**. **a)** Side view of the bulk crystalline structure of $MnBi_2Te_4$ with red arrows showing the interlayer AntiFerroMagnetic order. The crystal cleavage in this layered compound takes place at the van der Waals gap, thus exposing a Te layer of a Septuple Layer at the surface. **b**) Top and **c)** side views of the surface crystal structure. **d)** Atomically-resolved STM image of the surface of the cleaved sample (1 V and 0.3 nA) showing the hexagonal array of Te atoms and some defects (dark triangular depressions and a bright circular protrusion).

Ferrofluids and bio-ferrofluids: synthesis, structure, properties and some applications

Ladislau Vékás

Ferrofluids are ultrastable colloidal suspensions of magnetic nanoparticles, which manifest simultaneously fluid and magnetic properties. Their magnetically controllable and tunable feature proved to be an extremely fertile ground for a wide range of engineering applications. More recently, biocompatible ferrofluids produced a considerable increase of the applicative potential in nanomedicine, biotechnology and environmental protection. This talk offers a brief overview on the synthesis and advanced characterization of ferrofluids, bio-ferrofluids and ferrofluid based magnetorheological fluids. There will be summarized results of electron microscopy, vibrating sample magnetometry, Mössbauer spectroscopy, dynamic light scattering, small-angle X-ray and neutron scattering, and magneto-rheometry investigations revealing the similarities and differences between the three types of magnetically controllable fluids. The applications will be exemplified by leakage-free rotating seals operating in nuclear power units, MR flow controller devices in hydraulic machines, hybrid semi-active MR dampers for earthquake protection systems and by ferrofluid droplet splitting for antibody extraction processes.

Multifunctional therapies based on magnetic and photoactivated nanoparticles

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In biomedicine, magnetic and photo-induced hyperthermia therapies employ nanoparticles as heating sources for efficient therapeutic purposes in the fight of oncological diseases. However, these therapeutic effects have been also observed by localized nanoparticle heating without a detectable macroscopic temperature rise [1]. Local heating effects produced at the nanoparticle´s surface are a key issue to evaluate the onset of thermal doses and quantify possible side effects derived from achieving high local temperatures (i.e. destroying healthy cells in the tissue, degradation of proteins and enzymes in the extracellular medium, etc). In this work, we investigate the heating efficiency of nanoparticles subjected to magneto- and photo-thermal effects in combination with other therapeutic approaches methods in different biological environments [2,3]. Moreover, we also explore the use of the extended X-ray absorption fine structure (EXAFS) analysis as a direct and *in situ* probe to determine the local temperature at the nanoscale of magnetic and photoactivated nanoparticles upon laser photo-excitation, revealing significant nanothermal gradients [4].

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The magnetism of surface supported sheets and chains: Emergence of unprecedented properties by supramolecular control

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Supramolecular assemblies of molecules on atomically flat substrates represent a rich platform to investigate the interplay of fundamental interactions and the emergence of novel properties in low-dimensional materials. Here we demonstrate different architectural designs [1,2] leading, ultimately to the direct observation of long-range ferri-magnetic order emerging in a two-dimensional supramolecular Kondo lattice. [3] The lattice consists of paramagnetic hexadeca-fluorinated iron phthalocyanine (FeFPc) and manganese phthalocyanine (MnPc) molecules co-assembled into a checkerboard pattern on single-crystalline Au(111) substrates. Spectro-microscopy correlation experiments comprised by X-ray magnetic circular dichroism (XMCD) experiments and scanning tunneling microscopy and spectroscopy (STM/S) provide unambiguous evidence for the long range and local magnetic ordering. After magnetization at high magnetic fields (B > 6.5 T), remarkably, we find remanence in out-of-plane direction. First-principles calculations reveal that the FeFPc-MnPc antiferromagnetic nearest neighbour coupling is mediated by the Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction via the Au substrate electronic states. By chemical programming of the modules, different checkerboards were investigated containing metals of different oxidation and spin states.[4]

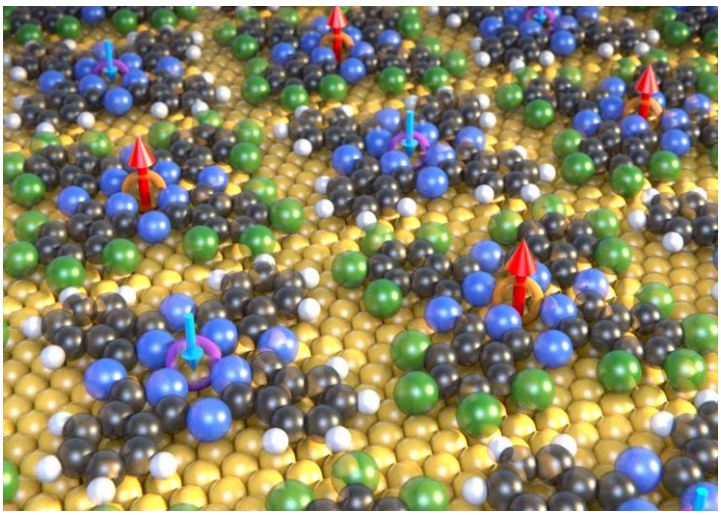

Fig. 1 FIG. 1: Model of the 2D phthalocyanine checkerboard with demonstrated ferrimagnetic order(LMN/SLS-XTREME) .

Realization of long-range magnetic order in surface-supported two-dimensional systems has been challenging, mainly due to the competition between short-range Kondo shielding and spin-stabilizing magnetic exchange interactions. By supramolecular engineering we provide a 2D spin architecture with a weak nearest neighbour and next nearest neighbour interaction. Such novel, tunable and configurable ‘quantum materials’ allow for novel and insightful experiments towards their application in quantum devices.

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Tuning dimensionality and type of magnetic order by auto-organization of Fe clusters in Fe-Au thin films and related spintronic effects

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Presently there is a large interest in studying specific magnetism and related magneto-functionalities in various configurations of magnetic nanoclusters, their field of interest spreading out from bio-medical applications (e.g. magnetic hyperthermia effects are very sensitive to their organization [1]) to spintronic applications [2]. A step forward is to conduct the organization of the magnetic nanoclusters in such a way to tune both the exchange interactions between the clusters as well as the local anisotropy, in order to make the analogy between atomistic magnetic elements in a crystalline lattice and similarly organized superspins. A full class of low dimensional magnetism and type of magnetic order, sensitive to both the magnetic dimensionality and superspin anisotropy of the system becomes available. If the magnetic clusters can be formed in a noble metal polymorph matrix, a large potential to tune further both the magnetic and electronic properties of the nanocomposite system may be achieved.
Fe_xAu_{1-x} nanophase thin films of different thicknesses (6 nm<t< 80 nm) and compositional ratios (0.1<x<0.7) are reported in this work. Complex morpho-structural, magnetic and magneto-transport investigation of the films were performed by X-ray Diffraction, cross-section Transmission Electron Microscopy, Magneto Optical Kerr Effect, Superconducting Quantum Interference Device magnetometry, Conversion Electron Mössbauer Spectroscopy and Magneto-resistance (MR) measurements. It was proven that depending on the preparation conditions and film thickness, different configurations of α-Fe magnetic clusters, from randomly distributed to auto-assembled in lamellar or filiform configurations, can be formed in the Au matrix. A close relationship between the Fe clustering process and the type of the crystalline structure of the Au matrix was underlined, with the stabilization of a hexagonal phase for x=0.3 and t>17nm. Due to different types of inter-cluster magnetic interactions and spin anisotropies, different types of magnetic order from 2D Ising type to 3D Heisenberg type, as well as superparamagnetic behavior of non-interacting Fe clusters of similar average size, were evidenced [3]. In plane magneto-conduction experiments have been performed with current flowing along or perpendicular to the magnetic easy axis (e.g. along or perpendicular to the lamellar or filiform configurations, simulating therefore both the CIP and CPP typical spintronic geometries) and with the magnetic field applied both perpendicular and transversal to the film. The obtained unusual MR effects were discussed with respect to specific magnetization reversals (associated to different types of magnetic order), evidenced by experiment and micromagnetic simulations, respectively.

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Spin asymmetry in the two-dimensional electron gas at $SrTiO_3$(001) surfaces

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$SrTiO_3$(001) perovskite single crystals are used as substrate for the synthesis of many perovskite materials by pulsed laser deposition (PLD) or magnetron sputtering. Recently, it was proposed that defects which might result in the population of Ti 3d states can promote electrons into the conduction band, such that these surfaces become conducting [1]. Such situations can be controlled by heating in presence or in absence of oxygen (in ultrahigh vacuum). If the sample is annealed in ultrahigh vacuum, it delivers oxygen, such that a part of the cations are found in oxygen depleted environments (i. e. locally suboxides are formed) and then the overall positive ionization state decreases. Also, irrespective of the sample stoichiometry, at surfaces of $SrTiO_3$(001) single crystals a two dimensional electron gas (2DEG) is formed [2]. Magnetic properties in oxygen depleted samples are reported since more than one decade [3], but to date no clear evidence was provided for spin asymmetry, nor any correlation was established with the presence of 2DEG.
On the other hand, according to a new criterion for band ferromagnetism proposed in Ref. [4], samples which start to show even a weak population of the 3d states of the transition metal should present spin asymmetry. The criterion mentioned above relies only of the shape of the density of states (DOS) in absence of spin asymmetry $g_0(\varepsilon)$ and is written as:

$$\left(\frac{dg_0}{d\varepsilon}\right)_{\varepsilon_F} \int_{\varepsilon_{min.}}^{\varepsilon_F} g_0(\varepsilon')d\varepsilon' < {g_0}^2(\varepsilon_F)$$

where ε_F is the Fermi level energy. If one approximates that near the bottom of the conduction band $g_0(\varepsilon) \approx \alpha\varepsilon$ and $\varepsilon_{min.} = 0$, then the above criterion is written as: $\alpha \times (\alpha {\varepsilon_F}^2)/2 < (\alpha\varepsilon_F)^2$, which is satisfied no matter of the value of the parameter α or of the value of the Fermi level, provided it is close enough to the bottom of the conduction band. If the electron gas is two-dimensional, then in the roughest approximation the DOS is nearly constant [5] and its derivative is zero; again, the criterion for occurrence of spin asymmetry is satisfied.
It follows then the need to test these predictions by using spin-resolved photoelectron spectroscopy from the valence band. This was performed by using the CoSMoS (combined spectroscopy and microscopy on a synchrotron) setup installed at the SuperESCA beamline at Eletra, Trieste, in the framework of a GdR (research group) partnership between NIMP and Elettra. The base pressure of the setup is in the range of 2×10^{-10} mbar. The $SrTiO_3$(001) single crystal was cleaned by Ar^+ sputtering at 500 eV ion energy and 10 □A sample current (on an area of about 1 cm^2), during 10 minutes. After this procedure, it was checked that the surface is atomically clean, without any presence of the C 1s level. The sample was then annealed in vacuum up to 900 °C and low energy electron diffraction (LEED) confirmed it has a good surface crystallinity, see Figure 1. The sample was characterized then by high resolution X-ray photoelectron spectroscopy (XPS) and spin-resolved valence band photoelectron spectroscopy. After this experiment, the sample was again sputtered, then annealed in oxygen atmosphere (5×10^{-7} mbar) by following in situ the increase of the O 1s signal and the disappearance of the shoulder attributed to Ti^{3+} in the Ti 2p signal. After this treatment, the sample was again characterized by high resolution XPS and by spin-resolved photoemission from the valence band.
Figure 2 presents some high resolution XPS data. The Ti 2p spectrum clearly shows the presence of a shoulder due to Ti^{3+} for the sample annealed in ultrahigh vacuum, which disappears by heating in oxygen atmosphere. At the same time, in the valence band spectra there is a non-negligible DOS near the Fermi level for the sample annealed in ultrahigh vacuum, which is absent for the sample annealed in oxygen. The stoichiometry analysis of the sample annealed in ultrahigh vacuum revealed an oxygen deficit of 0.2 per formula unit. Figure 3 presents the result of spin resolved valence band photoelectron spectroscopy. It can be seen that the spin asymmetry in the sample heated in ultrahigh vacuum (with presence of Ti^{3+}) is a robust signal, while the spin asymmetry is much lower, though not vanishing, for the sample heated in oxygen. In other words, the hypotheses of this work were confirmed. Additionally,

one can observe that the spin asymmetry signal is important in the region of important signal of the valence band, which originates mainly from the O 2p orbitals. This is, however, not surprising: the availability of states near the Fermi level induces the rigid shift of spin DOSs one with respect to the other, but the strongest signal is still expected to be provided by the region with important DOS [4].

Figure 1. Low energy electron diffraction (LEED) of the $SrTiO_3$(001) single crystal after heating in ultrahigh vacuum. The directions for the spin orientations along (ch1 – ch2) and (ch3 – ch4) are inclined at 45° with respect to this image.

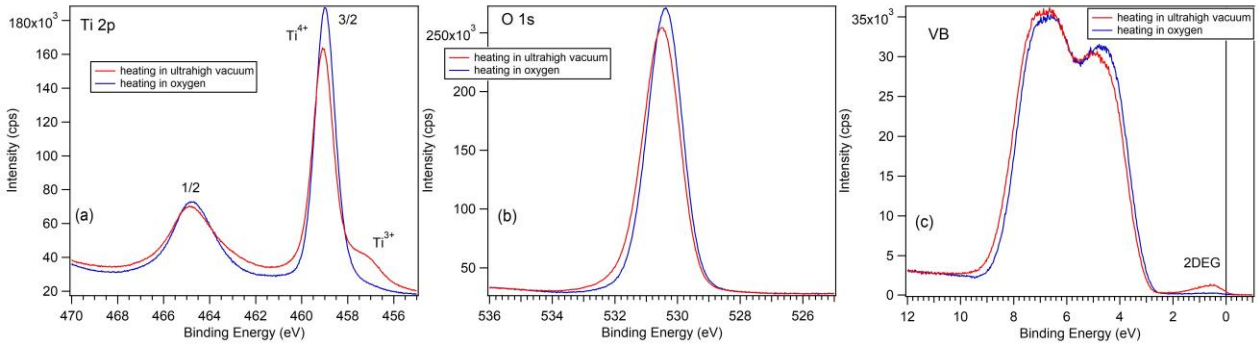

Figure 2. High resolution X-ray photoelectron spectra of Ti 2p (a), O 1s (b) and of the valence band (c). For (a, b) the photon energy employed was 650 eV, while for (c) it was 260 eV. Red curves represent sample heating in ultrahigh vacuum, blue curves represent the sample annealed in 5×10^{-7} mbar oxygen pressure.

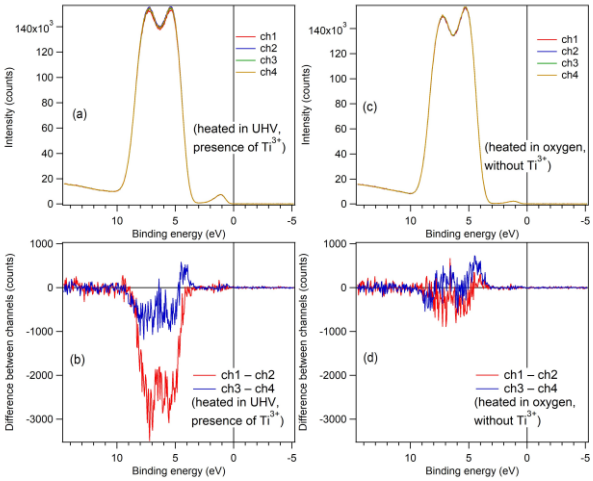

Figure 3 (a) valence band spectra obtained for the four spin channels with photon energy 100 eV, for the $STiO_3$(001) crystal annealed in ultrahigh vacuum; (b) differences between spin channels for that sample; (c) valence band spectra obtained in similar conditions for the crystal annealed in oxygen; (d) spin asymmetry for this crystal.

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Nanomagnetism applied to the development of sustainable permanent magnets for energy and transport applications

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Nowadays the lack of sustainable access to several Critical Raw Materials (CRMs) is probably the most serious challenge faced by the European raw materials sector with the permanent magnets to be one of the most vulnerable categories from this condition. Magnets containing CRMs (namely, rare earth-based magnets) are fundamental components in energy and transport applications.
Since the discovery of AlNiCos and ferrites in the middle of the 20th century, every new type of magnet has contained 20 wt.% or more of rare-earth elements. As a consequence, nowadays there is an enormous performance gap existing between ferrite and the strongest magnets (NdFeB), covered exclusively by rare earth-based magnets.
This presentation will focus on the most recent advances that we have done on two promising rare earth-free alternatives by efficient nanostructuring and controlled phase transformation: MnAlC alloy and improved strontium ferrite.
MnAlC is a good candidate for substituting bonded NdFeB. This may be achieved by maximizing the magnetization through the development of the only ferromagnetic phase of the system (τ-MnAl). However, an effort focused solely on obtaining pure τ-MnAl may compromise coercivity. Application of the self-developed "flash-milling" method (30–270s) has led to efficient nanostructuring and controlled phase transformation of gas-atomized MnAlC particles, resulting in a coercivity of 3 kOe (tunable up to >4.5 kOe) accompanied by a high remanence, used in the fabrication of MnAlC magnets [1,2].
In spite of the long history of hard ferrites, the possibility of improving their magnetic performance means an exciting scientific and technological challenge [3]. The trend over recent years has involved the use of CRM additives (e.g. La and Co), which brings again a lack of materials sustainability. Here it will be shown the possibility of developing improved Sr-ferrite powder with a high coercivity (> 6 kOe) through the combination of grain size refinement, induced microstrain and the creation of a multiphase system based on resources widely available in Europe. Moreover, the presentation will include two successful cases of application of nanoscience towards sustainability:

(i) Recyclability of Sr-ferrite residues generated in an industrial production line [4].
(ii) Additional use of the recycled material in 3D advanced manufacturing.

The EU H2020 project *PASSENGER* will be briefly introduced, as one of the largest international initiatives aiming at the industrial fabrication of rare earth-free permanent magnets (based on MnAlC and improved ferrite) for electromobility applications, through the participation of 20 centres (13 companies) from 8 European countries [5].

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Developing Rare Earth-free and Hybrid Permanent Magnets: from the Synthesis of Customized Composites to Additive Manufacturing

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Multi-material additive manufacturing (AM) is attracting much interest in many high technological sectors such as energy, transport, electronics, and medicine. The combination of AM with the synthesis of composites allows for designing and fabricating objects with complex shapes and tuned properties for reaching high performance [1]. For permanent magnets (PMs), a present challenge is developing magnets by AM with no geometrical constrictions, high filling factor (FF), and no deterioration of their PM properties during processing [2]. The industrial fabrication of PM/polymer composites is typically done by mechanical methods. We showed recently, and for the very first time, the possibility of producing a continuous PM-based filament (meters long) using as a precursor a highly loaded PM/polymer composite synthesized by solution casting [3]. This technique allows for a scalable synthesis of customized composites making possible the choice of the polymer according to the requirements dictated by the final application. Moreover, it is of large scientific and technological interest to consider rare earth (RE)-free PM alternatives such as improved ferrites, and the promising MnAl-based alloys [4].
In this work, different composites (PM particles/polymer) have been analyzed consisting on several PM materials: gas-atomized τ-MnAlC (Fig. 1(a)), Sr-ferrite, NdFeB, and hybrid (NdFeB/Sr-ferrite) powders embedded in different polymer matrix. The starting powders showed different mean particle size ranging from the 5 μm of Sr-ferrite to the 50 μm of NdFeB powders. MnAlC powders showed an intermediate mean particle size between 15 and 30 μm. This work will show the effect of particle size, polymer and fabrication parameters on the properties of the final products, showing that they are key factors to be considered and optimized for obtaining flexible and continuous highly loaded filaments.

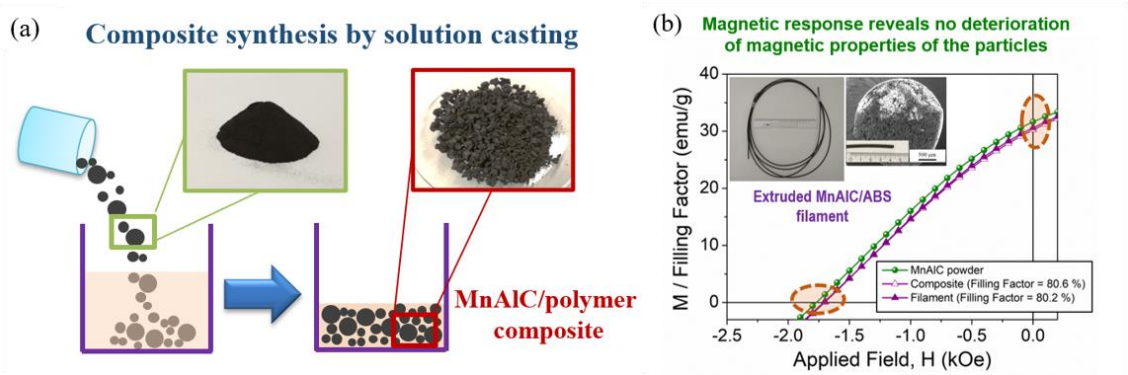

Fig. 1. (a) Composite synthesis by solution casting with representative images of the starting MnAlC powders and the resulting MnAlC/polymer composite. (b) Second quadrant of the room temperature hysteresis loops for the gas-atomized MnAlC particles, MnAlC/polymer composite and extruded filament. Inset shows an extruded MnAlC-based filament together with an SEM image of its circular cross section.
Particle size plays a key role when extruding the composites in order to obtain flexible filaments with a high FF [3,5,6]. It has been observed that if the mean particle size is larger than 20 μm (coarse particles), the extrusion process is affected by the composite rheology under the exerted pressures and, consequently, the FF of the extruded filament is reduced [5,6]. A key result from this study is the demonstration that, by mixing particles with different size and optimizing the fine-to-coarse particles ratio, it is possible to obtain flexible filaments with increased FF (up to 90%), i.e., leading to an enhanced effectiveness of the extrusion process [5,6].

MnAlC-based composites were synthesized by solution casting (Fig. 1(a)) making possible to tune the FF, reaching extremely high values above 85% [3]. Composites were extruded into continuous and flexible filaments with PM properties (Fig. 1(b)), with a length over 10 m [3,6]. The homogeneity of the composites and filaments has been determined by scanning electron microscopy, SEM (Fig. 1(b)). Processing of SEM images allowed for obtaining the FF of the composites and filaments. Vibrating sample magnetometry (VSM) has been used for accurately determining the FF of composites and filaments, being positioned as a faster technique in comparison with image processing. Moreover, and important in view of practical applications, this technique has demonstrated the no deterioration of PM properties of the starting particles after composite synthesis and filament extrusion processes (Fig. 1(b)) [3,5,6]. These results will be compared to those obtained in the fabrication of filament prepared from composites based on Sr-ferrite, NdFeB and hybrid composites containing fine (Sr-ferrite) and coarse (NdFeB) particles.

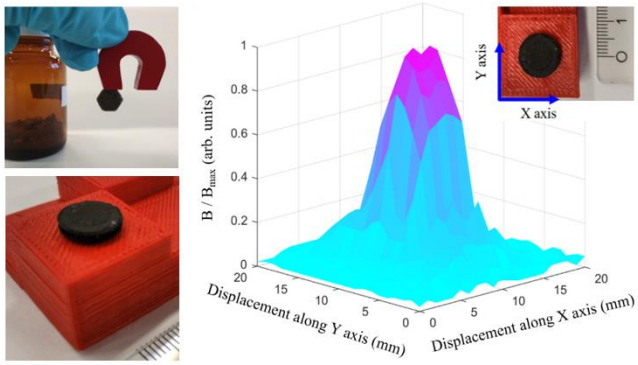

Fig. 2. Magnetic pieces obtained by additive manufacturing using an extruded filament based on MnAlC particles, together with the magnetic flux density measured at the surface of a 3D-printed disc.

Optimized MnAlC-based filament (with a high MnAlC content above 80 wt.%) was used for 3D-printing objects as a proof-of-concept (Fig. 2). The measurements of the magnetic flux density at the surface of the printed objects (Fig. 2) have proved that alternative PM materials can be efficiently synthesized and processed for developing novel PMs by AM under controlled processing temperature, which might be used in sensing devices [6].

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Nanocomposite permanent magnets based on Mn-Bi alloys. The role of LTP phase content on the structural and magnetic behaviour.

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With the fast development of green-energy technologies, the consumption of Nd-Fe-B permanent magnets (PMs) is expected to double or even triple in the near future, since they are the key components in the evolution of transportation technologies and renewable energies Moreover, today's modern applications are pushing the limits of the properties associated with conventional NdFeB-based PMs to operate at elevated temperatures. In particular, in automobile industry, the anticipated switch to full electric drive in the next 30 years will create a strong demand for magnets. For this reason, alternative solutions to the Rare Earth PMs are a huge issue nowadays and researcher all over the world are looking for cheaper and more reliable solutions.
MnBi alloys are one of the promising candidates for PM applications, since the MnBi low-temperature phase (LTP) has high magneto-crystalline anisotropy and a remarkable positive temperature coefficient of coercivity [1]. However, the encountered issues during the fabrication are driven by phase segregation effects and inhomogeneities that are difficult to control or eliminate, thus leading to low experimental reproducibility and uncorrelated results for structural and magnetic properties [2].
To overcome these issues, we propose an innovative method to increase the content of LTP by integrating an intermediary separation process during the manufacturing of Mn-Bi melt-spun ribbons. This method is suitable for obtaining Mn-Bi ribbons with high content LTP, as the removal of Mn excess from the alloy occurs during the ribbons formation through the melt spinning technique. As the method employs the removal of the excess unalloyed metals, the global weight yield is in range of 53-64 wt.%, but it can be increased by recovering the metallic elements and reprocess them. After a proper thermal treatment of the as quenched Mn-Bi melt-spun ribbons, over 99.0 wt.% is represented by the LTP phase with good saturation magnetization (~ 60 $A \cdot m^2/kg$) and coercivity (1.33 kOe).
We have also developed a new “exchange spring”- type system consisting of both Mn-Bi and Nd-Fe-B magnetic alloys. While the classical concept of “exchange spring” PM considers two magnetic phases with enhanced coercivity and B_s, coupled by exchange interactions, in our new approach the coercivity is “borrowed” from NdFeB, while MnBi enhances the thermal stability. The powders prepared from precursor melt-spun ribbons, were mixed in different ratios and compacted by spark plasma sintering (SPS) or cold die-compaction technique. The studies done on different compacts revealed that the cold pressed sample containing 90%wt. NdFeB and 10%wt. MnBi exhibits an enhanced “exchange spring" behaviour, with $H_c > 1.2$ T, $M_s \sim 120$ $A \cdot m^2/kg$ and the average temperature coefficient of coercivity, $\beta = 0.3812$ (%/°C) in the 298–423 K temperature region, representing a successful approach for our needs.

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Additive-induced phase stabilization in RE-free nanocomposite magnets

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Alloys having nominal composition $Mn_{53}Al_{45}C_2$ and $Mn_{52}Al_{46}C_2$ were prepared by the melt spinning method and were subjected to complex structural, morphological and magnetic investigations. As these alloys can exhibit tetragonal $L1_0$-type, τ phase, they have good potential as rare earth (RE) – free magnets. It is therefore important to monitor the $\varepsilon - \tau$ phase transformation, the stability and the magnetic features of the tetragonal phase in a whole temperature interval. By using synchrotron X-ray diffraction, it has been proven that the $\varepsilon - \tau$ phase transformation occurs gradually, with the τ phase becoming predominant, only after 450°C. Moreover, this phase has been proven to be quite stable without any grain growth even at the highest temperature investigated of 800°C. Low temperature behavior was thoroughly investigated using a complex combination of major and minor hysteresis loops combined with the zero field cooled – field cooled magnetization protocols (ZFC-FC). Two different regimes: blocking and superparamagnetic were documented. A spin reorientation transition was proven to occur at 55K while a maximum magnetization observed in ZFC-FC curves proved that at about 75K a ferro to antiferromagnetic transition occurs. The existence of a blocking regime, below 55K, that is characteristic to nanogranular systems with superparamagnetic behavior, has shown further development towards obtaining RE-free magnets.

POSTER PRESENTATIONS

Long-ranged Cu-based order at cuprate/manganite interface

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We present a resonant inelastic and elastic X-ray scattering (RIXS/REXS) study of epitaxial $YBa_2Cu_3O_7/Nd_{1-x}(Ca_{1-y}Sr_y)_xMnO_3$ heterostructures (NYN). We show that the Copper charge density wave (Cu-CDW) order of the near optimally doped $YBa_2Cu_3O_7$ layers can be strongly modified via the hole doping and tolerance factor of $Nd_{1-x}(Ca_{1-y}Sr_y)_xMnO_3$, i.e. by changing x and y.

At x=0.35 we observe a quasi-2D Cu-CDW order with d_{x2-y2} orbital character that resembles the one that is commonly found in strongly underdoped bulk YBCO. The strength of the corresponding Bragg peak at $Q_{\parallel}\approx0.3$ r.l.u. gets strongly enhanced as the tolerance factor of the manganite layers[1] is decreased and its CE-type antiferromagnetic and charge/orbital ordered (COO) is reinforced[1].

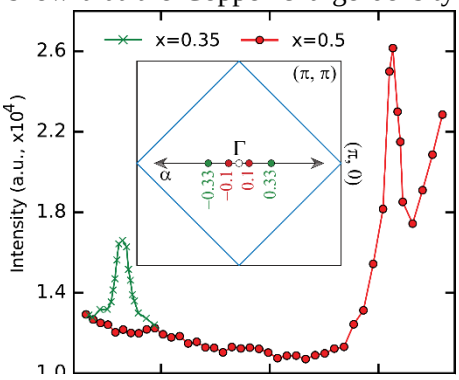

Figure 1: Comparison of RIXS measurements on manganite/cuprate heterostructures indicating the presence of the usual d_{x2-y2} Cu-CDW for x=0.35 and the new d_{z2} type Cu-CDW peak at $Q_{//}$=0.1 r.l.u. for x=0.5 (σ − polarization).

Upon increasing the hole doping of the manganite layers to x=0.5, we observe a new kind of Cu-CDW order which has a much smaller wave vector of $Q_{\parallel}\approx0.1$ r.l.u., a larger correlation length of about 40nm, and a different orbital character, i.e. d_{z2} rather than d_{x2-y2}, than the one commonly found in the bulk cuprates[2].

The origin of this new Cu-d_{z2} charge order is presently not understood, but seems to be rooted in the particular properties of the cuprate/manganite interface. The RIXS and additional x-ray absorption spectroscopy (XAS) data provide evidence for an important role of the orbital reconstruction of the Cu-ions at the interface with the manganite and a related transfer of electrons from the manganite to the cuprate. In particular, they show that the Cu-d_{z2} orbital of the interfacial Cu ions is strongly shifted up in energy and lies close to the Fermi-level such that it contains a significant part of the hole carriers, which usually mainly reside in the Cu-d_{x2-y2} orbital. This orbital reconstruction may well exhibit a lateral modulation along the interface that is linked with the anomalous d_{z2}-type Cu-CDW order.

While further studies are required to fully understand the interfacial coupling mechanism(s), the possibility of tuning the Cu-CDW holds great prospects for studying its relationship with high temperature superconductors and hopefully, for future quantum devices.

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Magneto-resistance effect in a ferromagnet/ insulator/superconductor heterojunction

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We present the effect of a trapped magnetic field in a base superconductor on magneto-resistive properties of a ferromagnetic layer. We have prepared in this respect by spark plasma sintering a MgB_2 superconducting disc with dimensions that enable trapping of magnetic field of about one Tesla unit at a temperature lower than the MgB_2 critical temperature. A 40 nm thick Fe layer (chosen as ferromagnetic layer) was deposited by magnetron sputtering on top of the MgB_2 disc with an intermediary MgO insulating layer of 100 nm in order to prevent a short circuit of the ferromagnetic layer in the superconducting state of MgB_2. This system was investigated comparatively with a reference one consisting of an Fe layer deposited in the same experimental conditions on oxidized Si substrate with MgO intermediary layer. A critical temperature of 37.5 K was inferred for MgB_2 from the zero-field cooled – field cooled curves. The magnetic and magneto-resistive properties were investigated below and above the critical temperature of the superconductor in order to emphasize the influence of the trapped magnetic field on the spin structure.

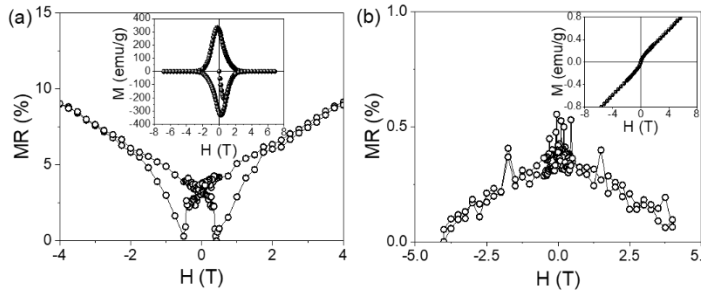

Figure 1. Magneto-resistance (MR) curve of S1_Fe collected at 15 K (a) and 40 K (b) in perpendicular geometry; insets present the magnetic hysteresis loops collected at the same corresponding temperatures. MR curves (collected with the field applied perpendicular to the sample plane) show not only an inverted evolution versus the applied magnetic field, but also more than one order of magnitude higher MR effect at the temperature corresponding to the superconducting state of the MgB_2. These features have been explained in the frame of an anisotropic magnetoresistance model, under the assumption that the orientation of magnetic moments is influenced not only by the external magnetic field but, at temperatures lower than the critical temperature of the superconductor, by the distribution of the trapped magnetic field which competes in addition with the intrinsic in plane uniaxial anisotropy of the ferromagnetic layer.

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Electrical-Magnetochiral Effect in ferromagnetic micro-coils.

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The discovery of magneto-chiral anisotropy in optics based on symmetry arguments raised the question if the same symmetry arguments can lead to new magneto-resistive effects in chiral conductors. When electrical conductors are helical, the resistance strongly depend on the current through, on the chirality and on the external magnetic field, all these parameters defining the Electrical Magneto-Chiral Anisotropy (EMCA) [1]. A current carried by a helical conductor will generate a magnetic field along the helicity, equivalent to a magnetization which direction depends on the direction of the current and the handedness of the conductor. When an external magnetic field is applied, it couples to the internal magnetization and affects the MR by its direction [2]. We propose the manipulation of shape and stress anisotropy, as well as of chirality in order to tune the MR properties of magnetic micro-wires. Glass coated $Fe_{80}Ni_{20}$ wire with diameter of 8.5 µm was fabricated by rapid ultrasolidification. An epoxy-encapsulated coil with 850 µm diameter and 250 turns was made of the wire. Spin configurations of micro-wire and micro-coil systems were studied by Transmission Mossbauer Spectroscopy (TMS) and Superconducting Quantum Interference Device (SQUID) magnetometry. MR properties (obtained by AC and DC Physical Properties Measurements System (PPMS)) were investigated comparatively for a straight micro-wire and for a micro-coil. TMS results indicate a spin configuration with average orientation of local magnetic moments outside the symmetry plane that contains the long axis of the coil. Anisotropic MR (AMR) effects are observed and in addition, in the case of the helical system, they are determined by electron scattering on specific magnetic configurations related to curvature-induced anisotropy. In the case of the coil system, AMR effects are evidenced also by angular dependent MR measurements. A linear EMCA was evidenced by comparing the electrical resistivity at opposite directions of the current flow: $\Delta\rho = (\rho_+ - \rho_-)$ at field values lower than 0.2 T where the rotational speed of the local magnetic moments is reduced with decreasing the field.

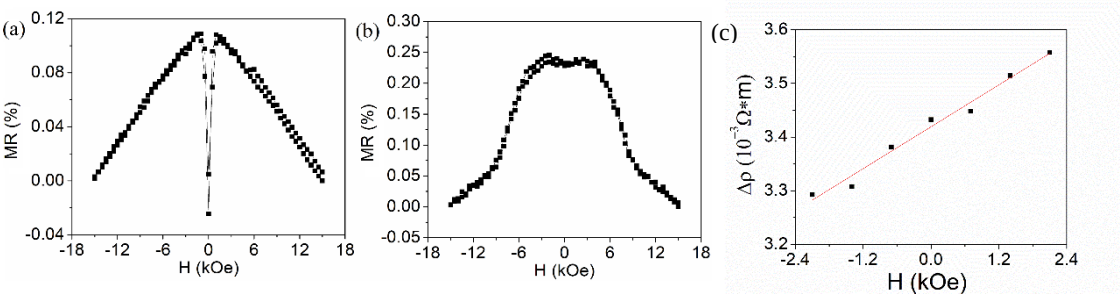

Figure 1. Room temperature MR curves of $Fe_{80}Ni_{20}$ micro-wire (a) and of micro-coil (b) systems in longitudinal geometry; highlight of EMCA in the case of $Fe_{80}Ni_{20}$ micro-coil (c)

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Magnetic-field-induced strain in NiMnGa Heusler-based ferromagnetic shape memory ribbons

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The ferromagnetic shape memory alloys (FSMA) applications are related to the well-known martensitic transformation (MT), a first-order phase transition occurring in the magnetically ordered domain. For Heusler type FSMA, the transition takes place between austenite (with B2 or ordered $L2_1$ structure) and either a seven-layer (14 M) or five-layer (10 M) modulated or a non-modulated (L10 tetragonal) martensite structure, depending on composition and thermal history [1]. The Ni-Fe-Ga near stoichiometric Heusler alloy with different substitutions has drawn much attention as an alternative to the brittle Ni-Mn-Ga FSMA [2]. In this work, we investigate the magnetic-field-induced strains (from thermal expansion and magnetostriction measurements in magnetic fields up to 5T) for Ni-Mn-Ga-Cu and Ni-Fe-Ga-Cu polycrystalline Heusler alloys prepared as ribbons from the melt-spinning technique [3-4]. All measurements were carried out on as-prepared rapid quenched ribbons and the thermally treated ones.

Information about magnetically field-induced strains was achieved from the linear thermal expansion measurements performed under cooling, at 5T. The applied magnetic field induces the nucleation of variants with an easy magnetic axis along the field direction as the sample is cooled down through martensitic transformation. The quenched-in stress stored in ribbons during the rapid quenching from the melt-spinning technique, and released by thermal treatments was found to influence the magnetic-field-induced strains of the ribbons. The highest strains were observed after the thermal treatment for Ni-Mn-Ga (3%at Cu) ribbons whereas, for the Ni-Fe-Ga (3%at Cu) ones, the thermal treatment reduces the strains values. The application of a magnetic field in the martensitic phase induces a weak motion of twin boundaries even in ribbons, and the magnetic-field-induced strain values obtained during heating are low, in concordance with saturation magnetostriction values. The magnetostriction behaviour may be explained by different mechanisms for the two types of samples: for the Mn-based ribbons is related to the twin boundary motion whereas for the Fe-based ribbons a magnetization rotation is involved.

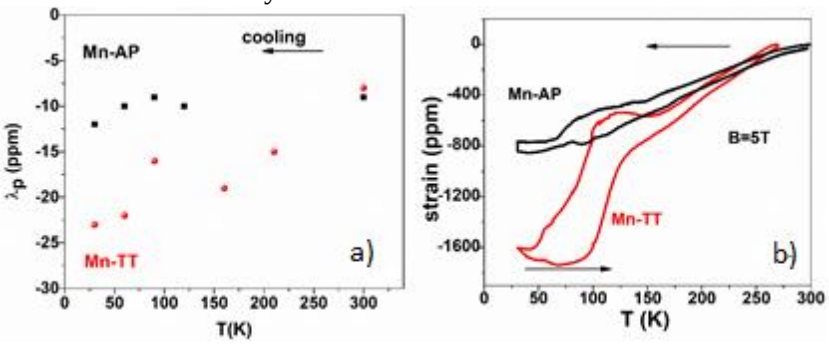

Fig. The temperature dependence of parallel magnetostriction (λp) (a) and thermo-strain dependencies recorded in an applied magnetic field (5T) (b) for as-prepared (Mn-AP) and thermally treated (Mn-TT) Ni-Mn-Ga-Cu ribbons.

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Rare earth garnet of $Gd_3Fe_5O_{12}$ type for future generation of electronic devices

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Ferrimagnetic materials close to the compensation point/temperature have attracted extensive attentions in spintronics due to their antiferromagnetic coupling, small net magnetic moment, fast domain wall moving speed, small stray field and less susceptibility to external field interferences [1-3]. Rare earth garnet ferrites (RIG) are materials with practical potential for design of magnetic switching devices due to their structural characteristics with a significant impact on their magnetic properties. This study proposes a complex analysis of $Gd_3Fe_5O_{12}$ nanoparticles obtained by our original technological route that combines a cheap and facile surfactant assisted hydrothermal method starting from non-ionic block copolymer Pluronic TMP123 environmentally friendly surfactant. Our results demonstrated the obtaining of a mesoporous structure, in a single phase with dimensions of about 16 nm. The room temperature Mössbauer spectrum of $Gd_3Fe_5O_{12}$ consists of a superposition of two Zeeman sextets assigned to the Fe^{3+} *a* and *d* sublattices of the ferrite garnet $\{Gd^{3+}{}_3\}_c[Fe^{3+}{}_2]_a(Fe^{3+}{}_3)_dO^{2-}{}_{12}$. The first sextet, of higher B_{hf}, is assigned to the octahedral 16*a* Wyckoff site, while the inner sextet accounts for the *24d* tetrahedral site. The hyperfine parameters are typical for $Gd_3Fe_5O_{12}$ [4] and the relative intensities of the octahedral and tetrahedral sublattices are in agreement with the theoretical cation distribution (2/3 ratio). At 5 K, the magnetization of saturation is about 89 emu/g. As temperature increased to 300 K, the magnetization decreased rapidly since the thermal fluctuation changed the main magnetic spin ordering. The temperature dependence of the spontaneous magnetization of the obtained $Gd_3Fe_5O_{12}$ reveals a compensation temperature (Tcp) of 286 K, much closer to room temperature as for previously reported systems.

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Magneto-functionalities in Fe-Gd ferromagnetic thin films close to the compensation point

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Amorphous Fe_xGd_{1-x} thin films of different compositions and thicknesses were prepared by magnetron sputtering and subsequently analyzed with respect to their magnetic behavior and related magneto-functionalities. A comprehensive set of samples were obtained where firstly the thickness is varied when different Fe/Gd elemental ratios are kept constant and secondly the compositions are varied for different film thicknesses which were maintain constant. The influences of these two parameters on the interplay between the two magnetic sub-lattices, one of Fe and the other of Gd, which are antiferromagnetically coupled were highlighted. The magnetization curves collected at different temperatures show the modification of the net contribution of the two magnetic sub-lattices by changing either the Fe/Gd elemental ratio or the film thickness.

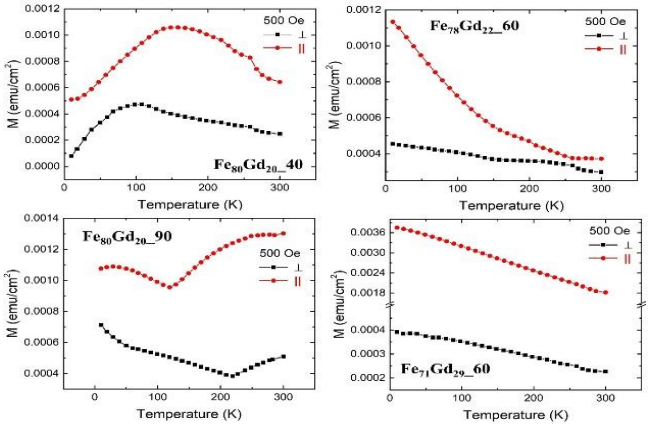

Fig 1: Magnetic compensation of sub-lattices depends on composition, thickness and direction of the applied filed with respect to the Fe-Gd thin films

^{57}Fe Conversion Electron Mössbauer (CEM) spectra give additional support to the specific magnetic behavior evidenced by magnetic measurements at Superconducting Quantum Interference Device (SQUID) magnetometry. Magneto-Optical Kerr Effect measurements revealed reversed hysteresis loops for sample compositions crossing the magnetic compensation point, showing the dominant effect of the Fe sublattice on the rotation of the light polarization vector. Complex shifted hysteresis loops were observed due to an atypical exchange bias behavior explaining the unusual magneto-resistance effects close to the compensation point.

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