EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Atomic scale properties of magnetic Mn-based alloys probed by Emission Mössbauer spectroscopy

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Abstract

Mn-based alloys are characterized by a wealth of properties, which are of interest both from fundamental physics point of view and particularly attractive for different applications in modern technology: from magnetic storage to sensing and spin-based electronics. The possibility to tune their magnetic properties through post-growth thermal processes and/or stoichiometry engineering is highly important in order to target different applications (i.e. MnxGa) or to increase their Curie temperature above room temperature (i.e. off-stoichiometric MnSi). In this project, the Mössbauer effect will be applied at ⁵⁷Fe sites following implantation of radioactive ⁵⁷Mn, to probe the micro-structure and magnetism of Mn-based alloys at the most atomic-scale. The proposed experimental plan is devoted to establish a direct correlation between the local structure and bulk magnetism (and other physical properties) of Mn-based alloys.

Requested shifts: 12 shifts, (split into \sim 3 runs over \sim 3 years)

1 INTRODUCTION/MOTIVATION

Manganese (Mn) is a 3d metal with electronic configuration [Ar]3d⁵4s². In recent years, several Mnalloys have been reported to exhibit room temperature (RT) ferromagnetism which has stimulated interest in terms of fundamental physics and applications. The aim of the present proposal is to understand in detail the physical mechanism behind the observed magnetic properties.

We propose to investigate three Mn-based alloys: (1) MnSi, where the stoichiometry (Mn/Si ratio) is deterministic of the magnetic properties, and defects seem to play a crucial role; (2) MnGa, where magnetic properties have been demonstrated to vary with thermal annealing and/or stoichiometry engineering; (3) Ni-Mn-X Heusler alloys, being of current interest due to multifunctional properties.

In all cases, the use of implantation of ⁵⁷Mn ($T_{\frac{y}{2}}$ = 1.5 min.) and ¹¹⁹ln ($T_{\frac{y}{2}}$ = 2.4 min.) for on-line ⁵⁷Fe and ¹¹⁹Sn emission Mössbauer spectroscopy (eMS) respectively, is proposed.

1.1 MnSi alloys

The implementation of spin functionality in silicon (Si), the mainstream semiconductor, is vital for establishing spin-based electronics with revolutionary potential in the modern information technology [Jansen12]. In this context, the synthesis and investigation of Si-based materials exhibiting RT ferromagnetism is of high interest due to the potentially easy integration into existing Si-based microelectronics. Moreover, transition metal-silicides are characterized by "unusual" electronic and magnetic properties, generating interest from the point of view of fundamental physics. Previously, thin film Fe silicide phases epitaxially stabilized on the appropriate substrates have been under intense investigation and RT ferromagnetism has been demonstrated in the non-stoichiometric c-FeSi phase (B2

Figure 1. Temperature dependence of magnetization for a MnхSi1-х sample (x ≈ 0.52) [Rylkov12].

structure) [Walterfang06]. The Mn_xSi_{1-x} alloy is another system of interest, which shows high temperature ferromagnetism when offstoichiometric, which cannot be adequately interpreted with available theories [Zhou10, Nikolaev09, Zhou09]. One of the realistic models [Men'shov11] describes the observed high temperature ferromagnetism in Si-rich Mn_xSi_{1–x} (x ≈ 0.35) alloys [Aronzon11] by the formation of $MnSi_{1.7}$ clusters [Zhou10, Zhou09] as well as the presence of defects with localized magnetic moments, leading to a strong increase in the Curie temperature (T_c) . Recently, it was experimentally found that T_c in Mn-rich Mn_xSi_{1-x} alloys (x \approx 0.52–0.55) is about an order of magnitude higher ($T_c \approx 300$ K) than that in the stoichiometric monosilicide MnSi

(B20 structure) ($T_c \approx 30$ K) [Rylkov12]- see Figure 1. Moreover, the deviation from the 1:1 stoichiometry, leads to a drastic increase in the mobility of the charge carriers, of about an order of magnitude at T \sim 100 K. It is believed that the existence of point defects with localized magnetic moments and/or nuclei of a different phase (such a B2 c-MnSi phase) explains the observed magnetic and electronic properties in Mn_xSi_{1-x} alloys (0.51 < x < 0.55). These models are supported by the results of numerical calculations performed within the framework of local-density-functional approximation [Rylkov12], but **the origin of ferromagnetism in non-stoichiometric MnxSi1–x alloys at RT is still not well understood**.

The on-line eMS measurements will provide information on the local environment of the ⁵⁷Fe probes at Mn sites, as well as the atomic-scale magnetic properties of Mn_xSi_{1-x} alloys. We aim to identify the role **of defects and/or inclusions of additional phases in the origin and evolution of the internal magnetic field in Mn-rich MnxSi1–x, as a function of composition x**. This will shed light on the source of the high-T ferromagnetism in this class of materials.

1.2 MnGa alloys

Rare-earth-free magnets such as MnGa have recently attracted much attention as new permanent magnets and materials for high-density perpendicular magnetic recording, magnetoresistive sensors, and spin-transfer-torque magnetic-RAM [Kurt11, Mizukami12, Zhu12, Zhu13]. MnGa alloys have been predicted to have spin polarization (i.e. the relative in-balance of spin up and down electrons at the Fermi level) up to 88% and $T_c = 730$ K [Balke07]. Magnetic films which display high perpendicular magnetic anisotropy (Ku) have several advantages when included in nano-scaled magnetic devices such as perpendicular magnetic tunnel junctions (p-MTJs), and the inclusion of MnGa in efficient p-MTJs has been reported recently [Ma12]. The ability to tune the combination of saturation magnetization values (M_s), coercive field (H_c), and magnetic damping constant (α) could open new possibilities in advanced devices. (001)-oriented single-crystalline MnxGa films have been synthesized by molecular beam epitaxy (MBE) on GaAs(001) [Zhu12, Zhu13]. Their magnetic properties: H_c, M_s, K_u, squareness of hysteresis loops (remanence M_r over M_s), magnetic energy product BH_{max}, and normal coercivity N_{cB} have been studied as a function of annealing time and temperature, Fig.2(a), and composition x in the range of $x =$ 0.76–2.6, Fig. 2(b).

Figure 2. Changes of M_S, M_r/M_s, H_c, H_{cB}, (BH)_{max} and K_u in Mn_xGa films, with (a) annealing temperature T_a (for 10 **min, open blue circles) and annealing time (at 450 °C, red points) for x=0.76, and (b) composition x, as taken from Ref. [Zhu13].**

Figure 2(a) evidences that Mn_xGa magnetic properties have little dependence on the annealing temperature *T*a (for 10 min. annealing), but remarkable changes are observed by changing annealing time at T_a = 450 °C (red points in Fig.2(a)). Figure 2(b) shows drastic variations of the magnetic properties with varying stoichiometry *x* from 0.76 to 2.6. For *x* = 0.97 – 1.75, the Mr/MS ratio exceeds 0.90, while a large decrease is measured outside this *x* range. The opposite trend is observed for the out-of-plane lattice constant *c*, as measured by XRD [Zhu13], which reveals a strong interplay between structure/strain and magnetic properties in this system. M_s gradually decreases from 450 to 52 emu/cc with increasing x, which has been attributed to an increasing antiferromagnetic coupling between Mn atoms [Zhu13]. The significant changes of H_c , seems to correlate with the changes K_u . The possibility to directly tune H_c, M_r/M_s, (BH)_{max}, N_{cB}, and K_u with annealing time is of extreme relevance. In-depth **understanding on the microscopic origin of these variations could suggest new routes towards optimization and/or engineering the properties of materials.** In this context, analysing the Mössbauer effect at Mn sites, as detected through the daughter ⁵⁷Fe atoms following implantation of radioactive ⁵⁷Mn* , will be highly important. The **main objective of the proposal is to establish a direct correlation between macro- and micro-scaled structural/magnetic properties in MnxGa materials**. Materials properties will be probed at the most atomic-scale by employing doses corresponding typically to 10^{-4} at.%, thus merely acting as "external probes" without varying the initial Mn_xGa stoichiometry. The samples to be investigated in the framework of the present proposal will be provided by the group headed by Prof. Jinhua Zhao (State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences), this group being one of the few groups first reporting compositional dependence of magnetism in MnxGa [Kurt11, Mizukami12, Zhu12, Zhu13]. The direct inclusion of this group in this proposal will certainly increase the potential impact of the planned experimental activity.

1.3 Other Mn-based alloys

In the framework of this proposal, a good opportunity exists to judge the feasibility of eMS to study more complex Mn-based ternary systems. In particular, Ni-Mn-X ($X = In$, Sn, Ga) alloys are attracting considerable attention due to the multifunctional properties (giant magnetoresistance, magnetic shape memory effect or large magnetocaloric effect) which arise as a result of the coupling between structure and magnetism [Yu06, Planes09, Chateerjee08]. The magnetic properties of this class of alloys (T_c of NiMnIn and NiMnSn is ~330 K and for NiMnGa it is ~400 K) are a consequence of a first-order martensitic transformation (MT) between different magnetically ordered structural phases. The MT occurs in the temperature range 250–300 K for all Ni-Mn-X alloys, and it can be easily tuned by the composition and/or thermal treatments. The "metamagnetic" character in X = In, Sn of the MT gives rise to new interesting phenomena (such as the magnetic-field induction of the MT, kinetic arrest of the martensite, exchange bias or the observation of a peculiar isothermal character in the MT), which are also being widely studied in recent years [Kainuma06, Ito08, Khan07, Pérez12, Pérez11]. The MT always takes place from a cubic austenitic phase, which shows the $L2₁$ Heusler crystal structure and nextnearest-neighbors atomic order. The degree of long-range atomic order may therefore affect both the MT characteristics and the magnetic properties of Ni-Mn-based alloys [Sánchez07, Recarte12, Sánchez08, Sánchez10, Sánchez11, Recarte12b, Sánchez12, Sánchez13]. The MT temperature, T_c, and M_s, all change with quenching in Ni-Mn-X (X = In, Ga) alloys [Sánchez07, Recarte12]. On the other hand, there is a null effect of quenching on the structural and magnetic properties in Ni-Mn-Sn alloys. This is thought to be related to the negligible $L2_1$ atomic disorder achieved with high-temperature annealing [Sanchez13]. Moreover, in Ni-Mn-Sn alloys there is an unexpected absence of L_{1} -B2 ordering transition, which also reflects the extraordinary stability of the L2₁ structure in this material. Therefore, even **though the structural order is the same, independently of X, the stability of the ordering depends on X, suggesting the importance of defects dynamics in determining the behavior of these alloys**. In fact, preliminary studies in NiMnGa [Merida10, Merida12] show the large role played by vacancy type defects in their behavior. The **influence of defects on the martensitic transformation and the defects recovery kinetics are still not understood in Ni-Mn-X**. A comprehensive investigation of the Ni-Mn-X (X = In, Sn, Ga) systems is beyond the scope of the present proposal. The aim will be to test the feasibility of the experimental methods proposed here to obtain information on the local structure and magnetism of this class of ternary alloys. We will focus on the Ni-Mn-Sn(In) system, with the main interest directed to characterize structural defect-complexes, the annealing of vacancy-type defects following ionimplantation, and to correlate this to atomic ordering and vacancy kinetics. The final goal is to determine the influence of structural defects on the characteristics of the MT, as detected (indirectly) through magnetoresistance and magnetocaloric effects. The results will be a strong basis to plan future proposals to INTC, with the aim of conducting a deeper investigation of ternary Mn-based Heusler alloys, with different X elements.

2 METHODS AND SAMPLES

2.1 Mössbauer spectroscopy

In this proposal, we want to make use of ⁵⁷Mn ($T_{\frac{1}{2}}$ = 1.5 min.) and ¹¹⁹ln ($T_{\frac{1}{2}}$ = 2.4 min.) that decay to the Mössbauer states of ⁵⁷Fe and ¹¹⁹Sn, respectively, to study the magnetic properties of manganese based alloys. The use of Mn is ideal to study these systems. After implantation, the 57 Mn adopts the lattice location of Mn during the 1.5 minute lifetime. After the β decay, the ⁵⁷Fe emission Mössbauer spectrum contains information on the magnetic hyperfine field that can be related to spin polarization of the Fe atom. There are mainly three reasons to apply implantation of short lived radioactive isotopes in this study:

1)) First, we can make use of site selective doping using Mn as the parent isotope. There is no chemical difference between the implanted species and the host material to be studied. This means that the obtained results are not compromised by possible different properties of probe/host material. In case of the Mn-Ga systems and some Heusler alloys, Indium should occupy different sites, giving alternative picture of the interactions with the neighbours.

2) Second, within the MnSi system and some Heusler alloys [Merida10, Merida12], the role of vacancy defects has been invoked in the discussion of the magnetic properties. Here, the eMS method, based on ion-implantation, is an ideal tool to study the role of defects in determining magnetic properties.

3) Third, to obtain a useful series of Mössbauer spectra, a concentration of only $\langle 10^{-4}$ at.% is needed. This low dose results in non-overlapping damage cascades that re-crystallizes at lower temperatures than amorphous layers, and in some cases (some metal systems) far below RT.

Usually, Fe atoms in 4–5 different configurations can be determined from eMS (depending on the complexity of the spectra), which is not possible with synchrotron Mössbauer methods.

For selected systems, the results obtained with ⁵⁷Mn implantations will be compared to Mössbauer spectroscopy of samples grown with stable ⁵⁷Fe and measured in home laboratories using commercially available sources of 57 Co ($T_{\%}$ = 272 days).

The Mössbauer collaboration at ISOLDE/CERN has access to implantation chambers suitable for on-line measurements using short-lived isotopes. Samples can be measured during implantation in the temperatures range from 90 K to ~1000 K. Angularly dependent measurements are possible (emission angles relative to sample surface normal 0–70°) and measurements in external magnetic field, up to 0.6 T at RT, and up to 0.3 T at temperatures below 530 K.

2.2 MnSi samples

 Mn_xSi_{1-x} alloys with different stoichiometry will be prepared by the Pulsed Laser Deposition technique (group of A. Zenkevich) using peculiar shadow geometry in a buffer gas to exclude the presence of droplets. The accurate control over the composition of Mn-Si thin film sample (within fraction of %), is achieved by the pulsed ("digital") nature of the co-deposition process from the elemental Mn and Si targets. The polycrystalline textured Mn_xSi_{1-x} samples \sim 2–10×10 mm² in size (depending on the required accuracy in composition) are grown on sapphire substrates kept at elevated temperature with composition in the range 0.4 < x < 0.6. Bulk single-crystal MnSi samples will be used as a reference. The as grown samples will be pre-characterized by Rutherford backscattering spectrometry and X-ray diffractometry to reveal the exact stoichiometry, thickness and the macroscopic phase composition

respectively. Temperature dependent Vibrating Sample Magnetometry (VSM) measurements will be performed to characterize "bulk" magnetic properties of as grown Mn-Si samples. Post-implantation characterization can also be planned.

2.3 MnGa samples

A series of MnxGa films with different Mn/Ga atom ratio x will be grown by molecular beam epitaxy (MBE) by following co-deposition of Mn and Ga at 250°C on 150 nm-GaAs-buffered semi-insulating GaAs (001) substrates, at base pressure less than 1×10^{-9} mbar. Samples will provided by the group of J. H. Zhao.

Compositions will be designed by controlling Mn and Ga fluxes during the growth, and checked carefully by high-sensitivity X-ray Photoelectron Spectroscopy (XPS) measurements before measurements at ISOLDE. Also, magnetization measurements will be conducted by superconducting quantum interference device (SQUID) before implantation with radioactive ⁵⁷Mn.

2.4 Ni-Mn-X alloys

Polycrystalline ingots of Ni_{50} -Mn_{50-y}-Sn_y (10 < y < 15) and Ni_{50} -Mn_{50-y}-($In_{x}Sn_{1-x}$)_y (13 < y < 17, x ~ 1) will be prepared from high purity elements by arc melting under protective Ar atmosphere (group of F. Plazaola). The ingots are re-melted several times. High-temperature thermal treatments (i.e. 30 minutes annealing treatment at different temperatures (ranging from 673–1273 K followed by quenching into ice water) can be performed to modify the atomic order and vacancies-defects content and to analyze its influence on the structural and magnetic properties.

3 EXPERIMENTAL PLAN AND BEAM REQUEST

For any material, the main tool is the usage of the ⁵⁷Mn-beam to record eMS spectra in a temperature series ranging from 90 to \sim 1000 K. The expectations are that at low temperatures, damaged environment dominate the spectra, and at high temperatures, defect-free environments are observed, which is reflected in the Mössbauer spectra (for example broadening of spectral lines). The temperature dependence of the spectra gives information on the role of defects and the annealing kinetics. Within each temperature series, the magnetic ordering temperature, and in the case of Ni-Mn-X alloys, also the MT transition will be accessible.

Each temperature series takes 2–4 hours to record, depending on the complexity of the spectra and beam intensity. In each system (MnSi, MnGa, NiMnX), we expect to start with the study of 3–5 samples (total of ~45 hours) to obtain the first general scientific overview. Some of this work will be exploratory, as very little is known about the behaviour of the materials under low fluence ion-implantation conditions.

Depending on the results obtained from the temperature series, samples will be selected for different measurements that we have available in our tool box:

- Measurements in external magnetic field. We can apply external magnetic fields up to 0.6 T at room temperature and in external magnetic fields up to 0.3 T below 530 K, and furthermore vary the direction between the γemission and the direction of magnetic field. Such experiments can give essential information that can be used to reveal the magnetic structure of samples.
- Quenching experiments (i.e. implantation at high temperatures, measurements at lower temperatures). Although not fully implemented in our setup, such experiments can be used to address the role of mobile defects.
- Time-delayed measurements, where spectra are recorded in time intervals after implantation [Gunnlaugsson09]. This is likewise useful to observe changing fractions due to instable defects created in the implantation process.
- At least in the case of MnGa systems, where we have access to single-crystalline material, angular-dependent measurements will be considered.

It is not unreasonable that a total of three shifts should be dedicated to this component of the proposal.

We also budget two ¹¹⁹In shifts to conduct ¹¹⁹Sn Mössbauer spectroscopy. This could be of interest in at least two respects. In the MnGa system, one expects In to occupy Ga sites, and this could give an alternative look at effects observed on the Mn sublattice with Mn implantations. The same applies for some of the Ni-Mn-X alloys. The ¹¹⁹Sn probe is more sensitive to magnetic hyperfine interactions and the monopole interaction (position of features in spectra) which depends on charge state/covalence in a simpler manner than in the case of Fe.

We furthermore plan ~15% additional beam time for calibration/contingency/opportunistic science/overhead, in order to address new scientific findings, not realised at the time of proposal submission. The investigation of the interplay between microscopic properties (this proposal) and macroscopic properties (characterization of materials) will be a steep learning curve. The long experience of the group in performing eMS at ISOLDE, demonstrates that sometimes important scientific aspects can be addressed (and sometimes solved), thanks to "last-minute changes" of the experimental plan, which follow inputs from fast visual inspection of spectra and preliminary analysis of the acquired data.

4 CONCLUSIONS/OUTLOOK

The use of ⁵⁷Mn radioactive beams to study Mn based alloys is a new exploration which is motivated by recent literature on interesting magnetic properties of Mn based alloys (see Section 1). Collaborations with leading groups already conducting research in this field will ensure access to materials of interest in this context. At least the following scientific questions will be addressed in this experiment

- The suitability of low fluence implantations for the study of Mn based alloys.
- Magnetic, structural, and electronic configurations of probe atoms.
- Kinetics of defect annealing.
- Role of defects in the observed magnetism.

If (or likely when) positive results are obtained on these questions, it is more than possible that this will result in new ideas for future research proposals/addendums.

SUMMARY OF REQUESTED SHIFTS:

^alf In RILIS is not available, surface ionization (W or Ta) can be a possibility.

5 BEAM DETAILS

⁵⁷Mn

⁵⁷Mn has been our work-horse for many years, decaying to the excellent Mössbauer state of ⁵⁷Fe (reasonable line-width and cross-section), and has been applied in many different scientific projects. In recent years, the Mössbauer collaboration has been able to use 5-6 shifts per beam-time without problems.

¹¹⁹In

In 2010 surface ionized beam (W) of intensity \sim (2-4)x10⁷ enabled spectrum to be recorded within \sim 1/₂ hour. In 2011, the ¹¹⁹In intensity was too low to justify using the beam. According to U. Köster (private communication, 2011) and V. Fedosseev (private communication 2013) the ionization scheme for In is a simple two-step process and should result in ~10 times more intensity. Moreover, the lasers can be optimized for the 2.4 minute isomer state of 119 In only.

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Appendix

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DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: (name the fixed-ISOLDE installations, as well as flexible elements of the experiment)

Please note that in January 2014 a new collection chamber should be delivered to ISOLDE to replace the existing GLM chamber. The safety file will be updated in due course once the new chamber arrives. The existing chamber will remain as a "spare" and will be adapted as a removable system allowing collections to be performed on the HRS separator.

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*