

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Emission Mössbauer spectroscopy of advanced materials for opto- and nano- electronics

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Abstract

In 2010, 15 shifts were awarded to the proposal entitled “Emission Mössbauer spectroscopy of advanced materials for opto- and nano- electronics”, for investigations on

(1) Paramagnetic relaxations in compound semiconductors

(2) Vacancy diffusion in group IV semiconductors.

On both subjects, major progress has been made, but after two years, many questions are still open, and we wish to propose a continuation of our investigations to derive solid conclusions on many aspects of these investigations.

Requested shifts: 17 shifts, (split into 1-4 runs over 1 year)



1 The IS501 experiment

In 2010, 15 shifts were awarded to the proposal entitled “Emission Mössbauer spectroscopy of advanced materials for opto- and nano- electronics”. The proposal focussed on four types of investigations

- (1) Paramagnetic relaxations in compound semiconductors
- (2) Vacancy diffusion in group IV semiconductors
- (3) Doping of Si-nano-particles
- (4) Investigation of phase change mechanisms in chalcogenides.

The INTC committee gave priority to the first two investigations, and based on the evaluation, (3) was dropped and (4) only taken as opportunistic science of low priority in our program.

In this addendum, we apply for beam time in 2012 to complete studies on outstanding questions on the two first subjects. In the following sections, the research fields are described, a brief status report after two years of the IS501 experiment is presented, and lastly we summarize our requirements to complete the picture(s) resulting from the investigations that have been done.

Details from beam reports and beam usage are given in section 8.

2 Paramagnetic relaxation in compound semiconductors

2.1 Introduction

Dilute magnetic semiconductors obtained by partial replacement of the cations in conventional semiconductors by transition-metal ions have been of major interest as potential innovative semiconductor-compatible magnetic materials for spintronic applications. Theoretical predictions of Dietl et al. [Dietl00] of room temperature ferromagnetism (RTFM) in ZnO (and p-type GaN) prompted a plethora of experimental studies of RTFM in ZnO and other metal oxides. Some experiments have observed RTFM (see e.g. [Jung02]) while others have not, and the origin of magnetism of transition-metal doped ZnO is controversial [Koji06, Osgur05] and There are inconsistent reports in the literature. Recently, the role played by defects in dilute magnetism have been discussed by many authors [Kapilashrami09, Khalid09, Wang08, Wang09], while others have suggested that the observed magnetic ordering may be due to unintentional precipitation, formation of secondary phases or misinterpretation of data [Potzger09, Zhou09].

By employing ^{57}Mn ($T_{1/2} = 1.5$ min.) to populate the Mössbauer state of ^{57}Fe , we have demonstrated that dilute Fe upon implantation occupies paramagnetic sites in ZnO and related model systems (MgO, $\alpha\text{-Al}_2\text{O}_3$ [Gunnlaugsson10a]). Using the ^{57}Mn beam at ISOLDE we are able to study the material at local Fe concentrations $< 10^{-4}$ at. % and any concerns about putative precipitation are obviated. If magnetic order exists in the ZnO, our results show that this has nothing to do with the dilute iron impurities. Representative emission MS results for ZnO in an external magnetic field are shown in Fig. 1.

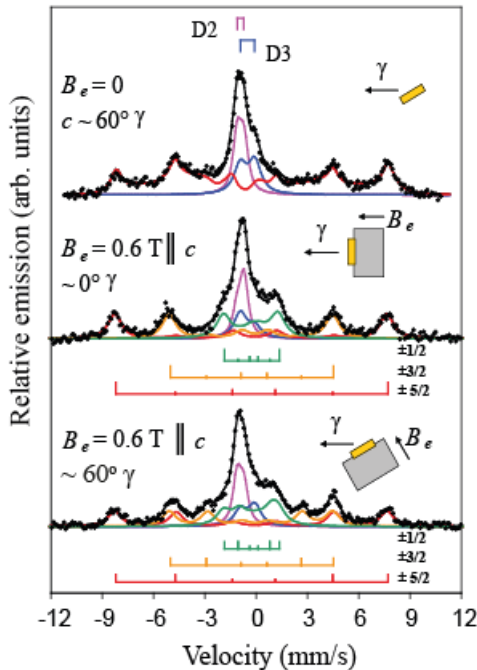


Fig. 1: Room temperature Mössbauer spectra of ^{57}Fe obtained after implantation of ^{57}Mn in ZnO. The spectra are obtained in ~ 0.6 T external magnetic field oriented as indicated with respect to the direction of propagation of the γ -quanta (adapted from [Gunnlaugsson10a]).

In the spectrum measured in external magnetic field at 60° , small features are seen at $\nu \sim -3$ mm/s and $\nu \sim 2.5$ mm/s that are clearly absent in the spectrum measured in magnetic field at 0° . This is characteristic for the disappearance of the lines belonging to the $\Delta m_l = 0$ transition for a $S_Z = \pm 3/2$ electronic state. Such a state can only exist in paramagnets (ferromagnets have only a spectrum resembling the spectrum from the $S_Z = \pm 5/2$ electronic state). This result shows that the electronic spin of the Fe^{3+} atom is not coupled to lattice defects from the implantation nor to the lattice, which would result in fast spin-spin or spin-lattice relaxations. The absence of such couplings shows that the Fe^{3+} atoms are unlikely to act as the seed needed for the proposed (long range) ferromagnetism.

2.2 Current status

As a part of the IS501 experiment, paramagnetism of Fe^{3+} has been verified in several model systems by measurements performed in an external magnetic field. Already published data includes MgO and $\alpha\text{-Al}_2\text{O}_3$ [Gunnlaugsson10b, Gunnlaugsson10c] (more data on several additional systems awaits publications). A method to extract the temperature dependent spin-lattice relaxation rates of Fe^{3+} in dilute systems has been developed [Mølholt10] which is sensitive to relaxation rate in the range of $\tau^{-1} = 4 \times 10^6 - 1 \times 10^8 \text{ s}^{-1}$. This method has been applied for MgO [Mølholt10], $\alpha\text{-Al}_2\text{O}_3$ [Gunnlaugsson10b] and ZnO [Mølholt12] (more data awaits publication). In this context, ZnO has shown some peculiar and interesting properties (cf. Fig. 2).

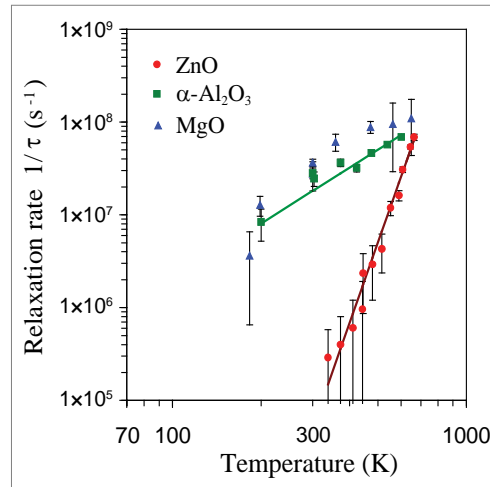


Fig. 2: Spin-lattice relaxation rates of Fe^{3+} obtained for the materials indicated (from [Mølholt12]).

The spin-lattice relaxation rate of Fe^{3+} in ZnO does not follow the T^2 dependence expected for temperatures $T > \theta_D/2$, where θ_D is the Debye temperature (see e.g. theoretical overview in [Mørup11] and [Srivastava80] and discussed in [Mølholt12]). Moreover, ZnO has a lower Debye temperature than MgO and $\alpha\text{-Al}_2\text{O}_3$ illustrating further this inconsistency with theory. The reason for this is at present unknown, but the experimental data has been provided to the scientific community [Mølholt12]. Preliminary analyses of paramagnetic relaxations of Fe^{3+} TiO_2 , SiO_2 , ZrO_2 , SrTiO_3 and GaN (unpublished results) also show the T^2 temperature dependence. These results require further investigation before publication. It should be emphasized that the study of these spin-lattice relaxation effects is only possible when the

local concentration of Fe is below 10^{-2} at. %. At higher concentrations, spin-spin relaxations start to play a role, and severely hamper the interpretation of the data. Therefore our studies at ISOLDE/CERN (employing probes at concentration $< 10^{-4}$ at. %) offers a unique opportunity to provide additional data to the scientific community on effects not explained by current theories on spin-lattice relaxation rates that are at dilution levels and temperatures that are outside the reach of any other experimental method.

Another peculiarity regarding ZnO is that the concentration of paramagnetic Fe^{3+} complexes increases with implantation fluence [Mølholt09], which may be due to implantation induced changes in the Fermi level, or due to pairing of Fe with structural defects. With good datasets acquired under different fluences, we have verified that the former explanation (implantation induced changes of the Fermi level) is indeed correct [Mantovan12]. ZnO is intrinsically n-type, and this feature suggests that the Fermi level moves toward the mid-gap under implantation conditions (implant isolation [Kucheyev03]). We have furthermore demonstrated that this change in the Fermi level is unstable at room temperature on a time scale of weeks [Naidoo12] and may contribute to explaining some inconsistencies in the literature regarding this material.

In 2010, a beam of ^{57}Co ($T_{1/2} = 272$ d) was developed at ISOLDE. Unfortunately, it contained high level of impurities (99% of the beam is stable ^{57}Fe), which hamper significantly the usefulness of this beam. In the case of ZnO, we were able to make use of the ^{57}Fe contamination in the beam as it allowed us to bridge the fluence gap between our ISOLDE data using ^{57}Mn (maximum dose 10^{12} cm^{-2}) and laboratory measurements utilizing stable ^{57}Fe ($\sim 10^{16}$ cm^{-2}) [Potzger06]. Measurements on a ZnO sample implanted with ^{57}Co ($5-6$) $\times 10^{12}$ cm^{-2} + ^{57}Fe ($5-6$) $\times 10^{14}$ cm^{-2} showed that Co/Fe enter damage sites of interstitial character (low Debye temperature) upon implantation fluence $> 10^{13}$ cm^{-2} [Gunnlaugsson12a]. It was moreover shown that this Fe in interstitial damage sites was mobilized with annealing at $T = 500^\circ\text{C}$ to form precipitates. Such precipitates would easily be detected with magnetization measurements, giving an alternative explanation of the observation of magnetism in these systems. Furthermore, these measurements resolved the apparent inconsistency in earlier Mössbauer data.

Having understood the role of the Fe^{3+} state in ZnO, the parameters of the damage component in ZnO and how to interpret slow paramagnetic relaxation, much more data are planned to be published soon. This includes: In-growth of the Fe^{3+} state [Mantovan12], annealing of the Fe^{3+} state [Naidoo12], damage in pre-implanted samples [Gunnlaugsson12b] and paramagnetic relaxation in other types of materials [Publication schedule to be determined].

Our results show that if diluted magnetism exists in any of the systems investigated, it has at least nothing to do with the 3d metal impurities. Some theoretical calculations suggest that defects may carry a magnetic moment [Wang08]. We tested this hypothesis by applying $^{119\text{m}}\text{Sn}$ Mössbauer spectroscopy following implantation of ^{119}In . The $^{119\text{m}}\text{Sn}$ probe is by factor of ~ 4 more sensitive to magnetic interactions than ^{57}Fe (see e.g. listing of the properties of Mössbauer nuclei in [MossOrg_Periodic]). The results corroborate the results obtained with ^{57}Mn that there is no strong magnetic interactions from nearby defects in ZnO, MgO and $\alpha\text{-Al}_2\text{O}_3$. However, an even stranger conclusion came from this work (cf. Fig. 3).

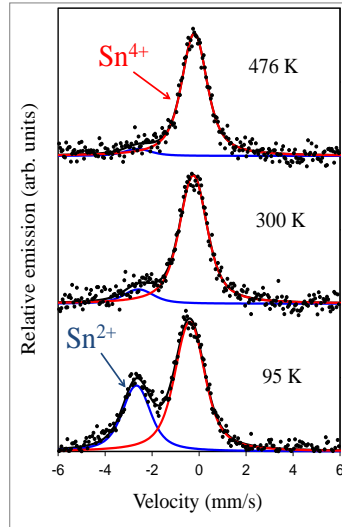


Fig. 3: ^{119}Sn Mössbauer spectra obtained after implantation of ^{119}In into a ZnO single crystal held at the temperatures indicated. The total dose is $\sim 10^{10} \text{ }^{119}\text{In}/\text{cm}^2$ per measurement.

At low temperatures, some of the Sn is observed in the expected 2+ state, but at elevated temperatures, Sn^{4+} is observed. This is in contradiction with the results obtained with ^{57}Mn , where one finds that above room temperature and for fluence $< 10^{11} \text{ cm}^{-2}$, Fe occupies regular lattice sites and is observed as Fe^{2+} by simple replacement of Zn^{2+} . Similar results were obtained in $\alpha\text{-Al}_2\text{O}_3$ and MgO. One straightforward explanation of this finding is that the relatively large In ion cannot simply exchange the metal ion, and hence is in a site with a charge compensating metal vacancy as the next nearest neighbour.

This result and its interpretation is of fundamental importance for several reasons:

- (1) ^{111}In is commonly used as a probe for perturbed angular correlation (PAC) experiments (see e.g. [Dogra09] and references therein). It is often assumed that the probe upon annealing occupies regular substitutional sites. Our results suggest that this assumption is not necessarily correct, and it would be of interest to be able to investigate this in more detail.
- (2) In + N co-doping of ZnO has been suggested as a method to obtain p-type doping in ZnO [Ye09]. If we prove that In implanted into ZnO does not lead to a simple replacement of Zn, it might help in a further theoretical and/or experimental endeavour to use such co-doping

Although the quality of the ^{119}In beam in 2011 was below usefulness, we were able to generate one spectrum after implantation in ZnSe.

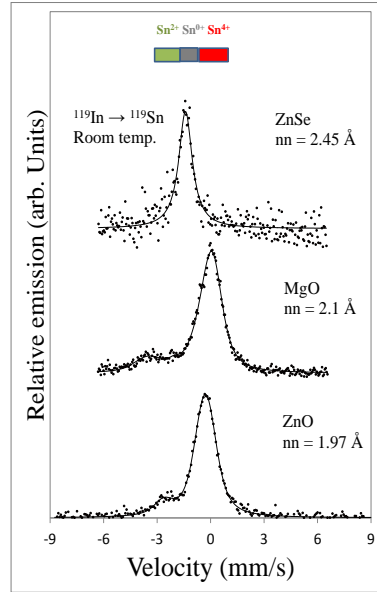


Fig. 4: ^{119}Sn Mössbauer spectra obtained after implantation of ^{119}In into the materials indicated, all held at room-temperature.

In MgO and in ZnO, one observes a broadened line due to Sn^{4+} , but in ZnSe, the probe atom enters a regular lattice site (no additional broadening). A possible, simple interpretation is that in MgO and ZnO the In atom is present on a site with a vacancy on the metal site..

2.3 Proposed methods and experimental plan

As a follow up on the results outlined above, we propose the studies described below which is a reasonable goal to reach within the one year before the 2013 shut-down.

- (1) What is the reason for the unusual temperature dependence of the paramagnetic relaxation rates in ZnO?
- (2) The lattice location/properties of In in oxides/group II-VI's

For (1), we plan to expand on our current list of oxides to obtain more data on slow paramagnetic relaxations of Fe^{3+} using implantation of ^{57}Mn . This would include temperature scans (90-600 K) in relevant oxides (e.g. SrO, BaO), selected compound oxides (BaTiO_3, \dots) and selected group II-VI semiconductors (Zn, Cd)(S, Se, Te). At least 5 different compounds would be investigated, and in selected cases angular dependent measurements in external magnetic field included together with time dependent measurements to document any possible metastable defects. Assuming “normal” intensity of the ^{57}Mn beam (usually $\sim 2 \times 10^8$ ions/s), recording of a reasonable high temperature series takes on the average 1 hour per sample, and 1.25 hour for a cold series. Good angular dependent measurements in external magnetic field take ~ 2 hour per sample. Thus for five different compositions, roughly 2 shifts would be required.

For (2), we propose using ^{119}In and $^{119\text{m}}\text{Sn}$ implantations. Here it is possible to make use of the dataset obtained in (1) because ^{57}Fe Mössbauer spectroscopy better determines the annealing reactions of implantation damage due to its higher sensitivity to quadrupole interactions. For ^{119}In , temperature scans (90-600 K) in some of the same types of samples

used in (1) should be performed. Assuming laser ionization and 4×10^7 $^{119}\text{In}/\mu\text{C}$ (see below), the total time needed for this part is (7-9 temperature steps, 20 minutes per step, in at least 5 types of materials) ~13 hours (1.6 shifts). $^{119\text{m}}\text{Sn}$ has not previously been applied to study these systems at all, so no data exists. According to the beam estimates ($\sim 1 \times 10^9$ $^{119\text{m}}\text{Sn}/\text{s}$, can be expected, see below), it will take roughly an hour to prepare the sample with 10^{13} cm^{-2} and an activity of 0.15 Mbq (4 μCi) which would be suitable for off-line Mössbauer measurements and PAC measurements. $^{119\text{m}}\text{Sn}$ should be implanted into at least 12 samples, among them ZnO, MgO, SrO, BaO, $\alpha\text{-Al}_2\text{O}_3$, SiO_2 , ((Zn, Cd)(S, Se, Te)). In the case of some of those (ZnO and others showing high damage resistance), higher doses should be applied and also implantations at elevated temperatures. Good suite of $^{119\text{m}}\text{Sn}$ implanted samples that can then be used in measurements during the long shut-down is obtained within 3 shifts.

2.3.1 Outlook

Upon completing the experimental plan (and off-line measurements) we will have obtained more data on paramagnetic relaxations of Fe^{3+} in oxides and compound semiconductors, giving unique dataset and pointers to the peculiar behaviour of Fe^{3+} in ZnO. Using this dataset, we will have documented the lattice properties of In/Sn in oxides and group II-VI semiconductors and answered specific questions such as whether In ions enter regular lattice sites upon implantation, whether it is the chemical nature of In that prevents it to enter regular lattice sites and whether it is possible to construct annealing procedures for $^{119\text{m}}\text{Sn}$ implanted samples in order to incorporate Sn into regular lattice sites.

3 Vacancy diffusion in group IV semiconductors

3.1 Introduction

Silicon has maintained its position as the most important material in semiconductor technology for decades and is probably the most studied material in the world. It is difficult to avoid transition metal ions completely during crystal growth. They have deep levels in the silicon bandgap and act as recombination centres. Transition metal impurities in general have low solubility, and are fast diffusers in silicon, and have received much attention in the literature (see e.g. review [Istratov99]). In previous experiments at ISOLDE we have concentrated on the physics of iron impurities in Si and other group IV semiconductors using ^{57}Mn (Experiments IS359 and IS426) (see [MossbColl]). In the present proposal we investigate some of the intrinsic properties of the silicon substrate making use of the implanted ^{57}Mn probe

Among fundamental open issues regarding the properties of silicon is the diffusivity and nature of the mono-vacancy at elevated temperatures. This behaviour of the mono-vacancy is a determining factor for the diffusivity of many species in the material, and understanding its behaviour can lead to better methods to model the material at processing temperatures. In this context, we have the unique situation that ^{57}Mn ($T_{1/2} = 1.5$ min.) decays to the Mössbauer state of ^{57}Fe giving the Fe atom recoil of average energy $\langle E_R \rangle = 40$ eV. This creates an interstitial Fe atom and a mono-vacancy in its vicinity. Some of the interstitial Fe atoms recombine with the vacancies at temperatures above 600 K, allowing us to study the behaviour of the vacancy at temperatures where no other technique can access such information.

Seeger and Chick [Seeger68] were the first to suggest that the mono-vacancies in silicon and germanium behaved differently. They suggested that the mono-vacancy in germanium was a point defect whereas the mono-vacancy in silicon was an extended defect, i.e. many Si atoms sharing the vacancy causing different diffusion behaviour at elevated temperatures. Bracht *et al.* [Bracht03], gave experimental evidence that the diffusivity of the mono-vacancy in Si at elevated temperatures (between 780°C and 872°C) occurs with much higher activation energy (1.7(5) eV), than observed at low temperatures (0.25-0.45 eV [Watkins91]). It was suggested that this behaviour is related to the extended nature of the vacancy at elevated temperatures, but their results are disputed [Suezawa08, Watkins08].

The hypothesis of a slow vacancy (high activation energy) at elevated temperatures has been supported by Mössbauer investigations at ISOLDE, using implantation of ^{57}Mn [Gunnlaugsson03, Weyer07], but further investigations are needed in order to substantiate these findings.

3.2 Status

The diffusivity of the (uncharged) mono-vacancy in silicon is outlined in the Arrhenius plot in Fig. 5.

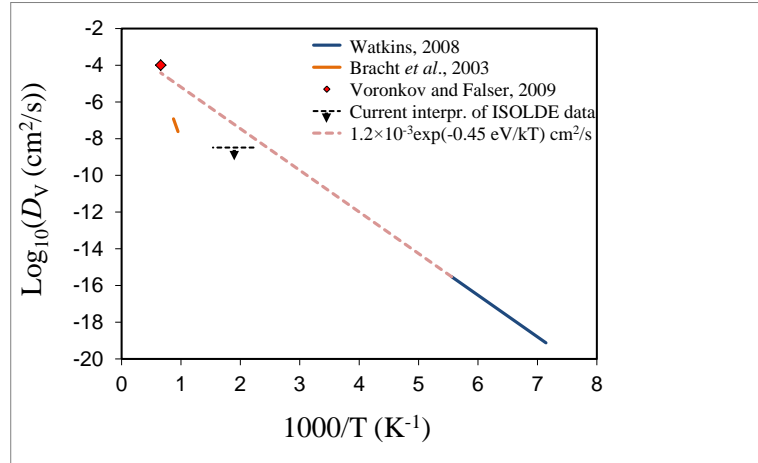


Fig. 5: Arrhenius plot showing the diffusivity of the mono-vacancy in silicon

Watkins *et al.* [Watkins91] found the mono-vacancy to behave as a point defect at cryogenic temperatures where it is thermally stable. Extrapolating his data to temperatures close to the melting temperature coincides with the data from Voronkov and Falster [Voronkov09] based on growth properties. Bracht *et al.*, [Bracht03] did not determine the exponential pre-factor of the diffusion but determined the activation energy (slope) to be much higher.

This is where we have the possibility to obtain unique data using implantation of ⁵⁷Mn for ⁵⁷Fe Mössbauer spectroscopy. Upon implantation and annealing of the implantation damage (~400 K) [Weyer03, Gunnlaugsson02b], the Mn probe is predominantly on substitutional sites and as Mn₁-V complexes, based on preliminary analysis of EC experiments [Wahl11]. In the recoil, a sizeable fraction of the Mn atoms is expelled into interstitial sites [Gunnlaugsson02a], creating a vacancy nearby. If the vacancy follows the mobility assumed by extrapolation of the Watkins data (dashed line in Fig. 5), one would expect the recombination of the interstitial Fe atom and the vacancy between 350–400 K. However, this recombination is only observed at 600–650 K, suggesting a “slow” vacancy. Moreover, the interstitial Fe atom does not jump into the vacancy to form substitutional Fe, but forms a metastable pair, possibly due to the extended nature of the vacancy [Seeger68].

It is unlikely that it is the presence of the interstitial Fe atom that affects the configuration of the vacancy in silicon. In SiGe (unpublished data from the IS426 experiment, awaiting more detailed data from current experiment), we observe a transition from a “slow” vacancy in Si_{1-x}Ge_x for $x < 0.1$ to a “fast” vacancy for $x > 0.25$.

3.3 Methods

The basic tools used by us, are temperature mapping and so-called time-delayed measurements (TDM) [Gunnlaugsson09] using ⁵⁷Mn. The temperature mapping allows us to make a firm identification of (overlapping) spectral components (position in spectra, quadrupole splitting, line-width, Debye temperatures) due to the different in-growth and disappearance of spectral components as a function of temperature. As the behaviour of some of the spectral components differs between materials, cross correlation of spectral parameters between different types of materials will make this identification stronger (the aim of the current proposal). In TDM we implant for relatively short time (~100 s) and follow the fate of spectral components as a function of time during the 1.5 minutes lifetime of ⁵⁷Mn. This

		Fe _I -V formation is not possible	observations are at 600 K	details
2	Ratio of Mn _S /Mn _I (-V)	Important input for determining what happens during the recoil		Emission channeling in the temperature range from 300-500 K
3	What is near Tetrahedral Mn _I	In MS we observe the ideal T site, while in EC it is shifted from the ideal T site	Mn _I forms a complex with vacancy. With a binding energy ~1 eV, it dissociates in the recoil and Fe _I is observed as ideal interstitial site	EC at low temperatures; measurements at T's where V is immobile should give the ideal interstitial sites
4	Assignment of Fe _S -V	Currently very tentative	Dissociation of VO pairs	MS Measurements in FZ and HP materials
5	Formation of Fe _S -V	Why is Fe _S -V only observed at high temperatures	Formed, but dissociated due to the recoil? Not enough vacancies around?	Comparison of MS results in all types of materials + EC at 500-750 K
6	Outdiffusion of interstitial Mn	Should lead to formation of new spectral lines in MS, some changes observed at ~800 K of unknown origin		EC Measurements on EC on ⁵⁷ Mn at T = 750-850 K or ⁵⁶ Mn, MS measurements at high temperatures in n ⁺⁺ Si

3.3.1 ¹¹⁹In

In 2010, we attempted to apply ¹¹⁹In ($T_{1/2} = 2.1$ min) for ¹¹⁹Sn Mössbauer spectroscopy. Among the aims was to apply TDM's to observe the formation of In-V pairs. Such pairs have been observed in ¹¹¹In PAC studies [Deicher87]. Surprisingly, the analysis of the data did not require the presence of a In/Sn-V component. A possible explanation for this is that it is too small to be observed, or that the parameters of such a component are too similar to the substitutional Sn to be distinguished from it with the statistics that were available.

To obtain a complete picture of the group IV semiconductors, implantations into diamond, SiGe and Ge were also performed in 2010. Fig. 7 summarizes some of the findings.

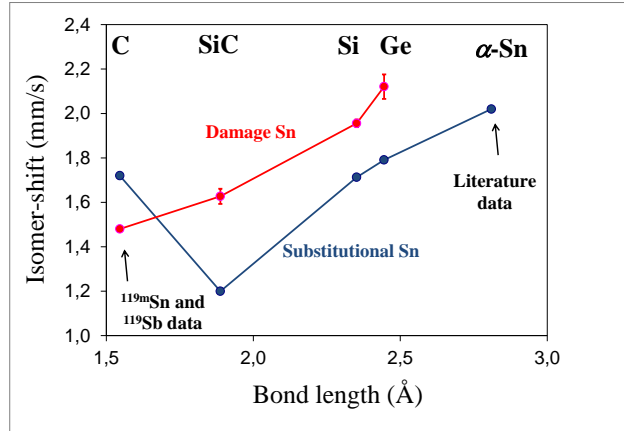


Fig. 7: Isomer-shifts of damage sites and in substitutional sites in diamond-type lattices of group IV semiconductors.

While SiC – α -Sn shows the expected pressure dependence of the isomer-shift for the substitutional line, there is an apparent jump for diamond (C). This is possibly due to the large size of the Sn atom, not being able to enter a simple substitutional site, and may be related to the same effect as observed in oxide semiconductors described in section 2.2. In 2010 we had only small diamond samples and were unable to get useful Mössbauer signal (which is strange in view of the fact that diamond has the highest Debye Waller factor of the group IV semiconductors). In 2011 we had a large area CVD diamond sample, characterized the annealing reactions with ^{57}Mn implantations, but the ^{119}In beam was too poor to allow for this data to be taken.

3.4 Experimental plan

The data taking for the tasks labeled as “very important” and most of the “important” outlined in Fig. 6 is estimated to take ~32 hours (~4 shifts) and this would cover items 4 and part of items 5 and 6 outlined in Table 1. The time needed for performing emission channeling on ^{57}Mn is based on experience from 2011 (items 2, 3 and part of 5 & 6). Here it was found that measurement at single temperature takes ~2 hours, so full mapping of two different types of samples is estimated to need ~24 hours (=3 shifts) [Wahl11]. The EC dataset obtained using ^{57}Mn ($T_{1/2} = 1.5$ min) will also help in the interpretation of the dataset obtained using RT implantation and annealing experiments with the longer-lived ^{56}Mn ($T_{1/2} = 2.6$ h) which is a part of the IS453 experiment [Wahl11]. It should be noted here that the short-lived ^{57}Mn is essential if one wants to study the lattice location of Mn as a function of **implantation temperature**. Such experiments cannot be performed with ^{56}Mn in reasonable time since after each implantation and measurement step one would have to wait about 6 h until the old activity has decayed and a new implantation at a different temperature can take place.

With laser ionization of In, it should be possible to further apply TDM’s in silicon to see whether In-V pairs form, and in that case investigate this capture of vacancies further, and to complete the picture of group IV semiconductors by temperature mapping in CVD diamond. One shift should be sufficient for this. This data could be supported by $^{119\text{m}}\text{Sn}$ implanted samples, where less than a whole shift would be sufficient.

4 Experimental and samples

4.1 Mössbauer spectroscopy

The Mössbauer collaboration has a multi-purpose implantation chamber to perform on-line experiments (cf. Fig. 8).

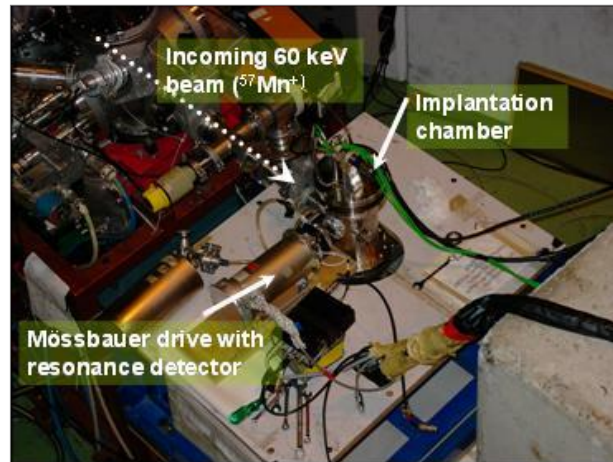


Fig. 8: Picture of the implantation chamber used for Mössbauer measurements of short lived isotopes in configuration used for elevated temperatures. The sample holder is attached to the lid and different lids are used for the different applications.

Different experimental conditions (temperature, rotation, external magnetic field) are installed by placing different types of lid's onto the chamber. Some of these facilitate up to 4 samples in the same load. With new pumps, sample change takes of the order 15 minutes. For the experiments we have detectors with ^{57}Fe enriched stainless steel electrodes and for the 2012 beam time a newly developed $^{57}\text{FeAl}$ detectors which we expect (hope) will increase the counting effect by factor 2.

The Mössbauer collaboration counts currently 16 researchers from 9 institutes (cf. Appendix 3). On the average 10-14 persons are active in data taking during the on-line experiments, ensuring enough manpower.

4.2 Emission channelling

The emission channelling group is led by U. Wahl. They have on-line chamber for performing emission channelling on short lived isotopes. During the 2011 beam time, a test using ^{57}Mn was performed and was successful. It was seen that sufficient data under single emission angle was obtained in ~40 minutes. Full measurement at single temperature (3 emission angles) takes ~2 hours, and these numbers have been used as the input in the beam request.

To illustrate the usefulness of ^{57}Mn for emission channeling, Fig. 9 shows the emission patterns obtained from room temperature implantation of ^{57}Mn in to n-type Si.

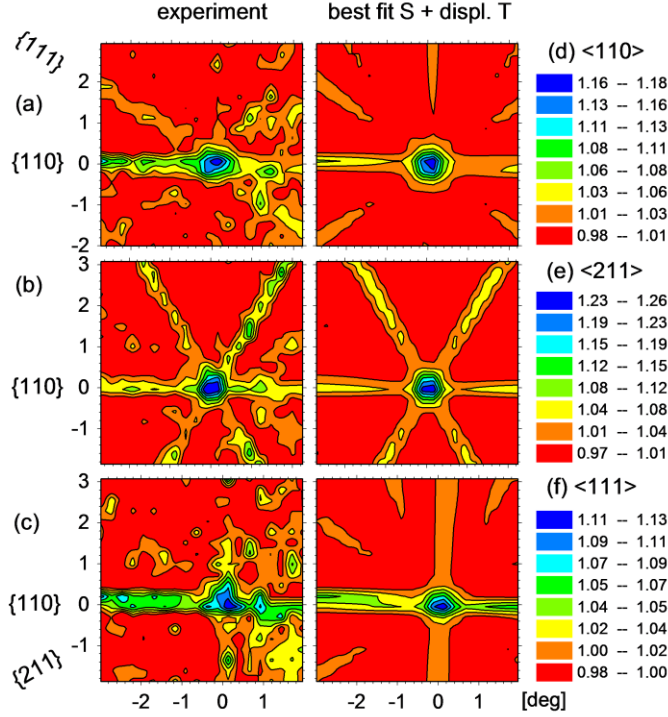


Fig. 9: Emission channeling and best fit, assuming substitutional site (S) and displaced tetrahedral interstitial site (T) [Wahl11]. Note there is a higher background toward the lower-left in the experimental data.

The fractions found are 12(2)% at S sites + 8(2)% displaced 0.4(2) Å from the ideal T-site. These fractions have not yet been background corrected, but illustrate that despite the relatively high electron energy that ^{57}Mn is a good EC isotope.

4.3 Perturbed Angular Correlation

It is possible to perform $e^- \gamma$ Perturbed Angular Correlations (PAC) experiments on $^{119\text{m}}\text{Sn}$ [Soares73]. It is an excellent PAC 3/2 isotope where K or L conversion electrons from the first transition are used for triggering the start while the M1 γ ray used in Mössbauer experiments triggers the stop. A Siegbahn-type spectrometer, providing measurements from 30 K up to 800 K, is in operation at ISOLDE and is the ideal setup for performing such measurements. Samples would be prepared for both PAC and Mössbauer spectroscopy, allowing for direct comparison between the results obtained by both techniques. Measurement times are similar as with Mössbauer spectroscopy (~weeks). In case that the Mössbauer results are complicated, PAC may give complementary information on their interpretation by determining values of the quadrupole interaction. Decisions on which samples would be potentially used for PAC will be taken on the basis of the Mössbauer results.

4.4 Theoretical calculations

Our identification of defects can be supported by theoretical calculations of hyperfine parameters. We collaborate with A. Svane at the Aarhus University in this respect. He applies quantum mechanical calculations based on the local density approximation to density

functional theory on a supercell with Fe or Sn atoms. The calculations give information on total energies and Mössbauer hyperfine parameters. Currently Svane is working on (among other things)

- (1) Calculations of isomer-shifts of substitutional Sn in diamond and Sn in a di-vacancy in diamond. This will become relevant in the interpretation of the “jump” shown in Fig. 7.
- (2) Calculations of isomer-shifts of Sn in oxides and II-VI's. This is motivated by the observation of Sn^{4+} instead of Sn^{2+} in several oxides following implantation of ^{119}In . In some of the II-VI's (e.g. ZnSe) the covalent nature of the bindings result in isomer-shifts that are not necessarily directly characteristic for the charge state of the Sn atom, and only with the aid of theoretical calculations one can come to firm conclusions.
- (3) Calculations of hyperfine parameters and properties of Fe in silicon. At the moment we have only very tentative identification of the $\text{Fe}_\text{S}\text{-V}$ complex, and this hypothesis can be supported or rejected by the results of theoretical calculations.

4.5 Samples

We have access to all sample types discussed in our proposal, in particular different types of Cz silicon single crystals from collaboration with the semiconductor group of Arne Nylandsted Larsen at Aarhus University in Denmark, where we also have access to moderately doped float-zone (FZ) grown material and some higher doped high purity (HP) MBE grown material. Oxides and selected II-VI's are available commercially and large area CVD diamond sample is available.

5 Summary of requested shifts

First, here is a list of how we intend to use the beam time (if awarded)

Method/Theme	Isotopes					Total
	⁵⁷ Mn	⁵⁷ Co ^a	¹¹⁹ In	¹¹⁹ Sn	¹¹⁹ Sb ^a	
1. Paramagnetic relaxations in compound semiconductors	2		1.6	3		6.6
2a. Vacancy diffusion in Si based semiconductors (Mössbauer shifts)	4		1	0.75		5.75
2b. Vacancy diffusion in Si based semiconductors (Emission channelling shifts)	3					3
Calibration (~10%)	0.25		0.15			0.4
Contingency/opportunistic science (~20%)	0.25	0.5	0.5	0.25	0.25	1.25
Total	9.5	0.5	2.75	4	0.25	17

^a Only used if other users are requesting suitable target/beam

This totals to 9.25 on-line Mössbauer shifts (⁵⁷Mn and ¹¹⁹In), which is similar to the shifts we have been able to use per year. Implantation of the other isotopes does not require the same manpower, as the Mössbauer measurements are performed off-line at home laboratories or at ISOLDE.

We request additional beam-time for contingency/opportunistic science. We often get requests for measurements of high scientific value, that require only 1-2 hour of beam-time. Such experiments have previously resulted in new proposals or allowed us to demonstrate that the physics program is feasible. In 2011, this addition was very important due to the low intensity of the beam and allowed us to get data for the main subjects of the proposal.

Table 2: Formal beam request.

Isotope	Minimum Intensity/ μC	Energy	Shifts	Target	Ion source	Desired Proton Current
⁵⁷ Mn (1.5 m)	$(2-3)\times 10^8$	≥ 50 keV	9.5	UC _x	Mn RILIS	2 μA ^a
⁵⁷ Co (270 d) ^b	9×10^7	≥ 50 keV	0.5 ^d	ZrO ₂ or YtO ₂	VADIS/RILIS(?)	2 μA (at beginning of run)
¹¹⁹ In (2.1 m)	$(2-3)\times 10^8$	≥ 50 keV	2.75	UC _x	In RILIS	2 μA
^{119m} Sn (292 d)	1×10^9	≥ 50 keV	4	UC _x	Sn RILIS	2 μA
¹¹⁹ Sb ^c (38 h)	4×10^8	≥ 50 keV	0.25 ^d	UC _x	Sb RILIS	2 μA
Total			17			

^aFor the Mössbauer part. > 0.5 μA for the Emission channelling part

^bWe take all nuclei that eventually decay to ⁵⁷Co. Note, ⁵⁷Co builds up within ZrO₂ or YO targets. By keeping these targets cold during proton irradiation enables long-lived ⁵⁷Co to be extracted by heating the target unit. Protons are not necessarily needed at this stage.

^cImpurities should be below ~10%, Tellurium impurities (¹¹⁹Te ($T_{1/2} = 4.7$ d)) have to be avoided.

^dIf suitable beam is provided for other purpose

5.1 Beam details

For all the isotopes, and especially the Mössbauer part of ⁵⁷Mn, it is requested that as close to 2 μ A of proton current be delivered as is possible. These runs should be dedicated to GPS only. Unlike some solid state runs, sharing with HRS in parallel is not feasible and results in lower yields (as in 2011).

5.1.1 ⁵⁷Mn ($T_{1/2} = 1.5$ min.)

This is the beam the group is very familiar working with. The measurements are done online in the setup described in section 4.1. Our experience for the last many years is that we use roughly 5-6 shifts per year of ⁵⁷Mn assuming “normal” intensities ($\sim 2 \times 10^8$ ⁵⁷Mn/s). In 2011, the intensity was much lower (see section 8).

5.1.2 ⁵⁷Co ($T_{1/2} = 270$ d)

⁵⁷Co is of high interest for the Mössbauer collaboration as the number of facilities offering this beam has decreased significantly since the 90’s. Unfortunately, the current beam provided at ISOLDE has high ⁵⁷Fe impurities to be well useful in all context. Experience from 2010 and 2011 (YtO₂ target and VADIS ion source) tells us that with optimum settings a weak source can be produced in half shift.

In principle, a RILIS scheme exists for Co beams, which would reduce the Fe impurities which have caused problems with these beams over the last couple of years. Although unlikely to be used during a “normal” ZrO₂ or YO run, this could potentially be employed at the end of the running period in 2012, if long-lived isotopes are being delivered instead of the usual separator courses. As mentioned above, protons would not be needed if the target had been already irradiated, but kept cold.

Implanted samples would then be shipped to home institutes.

5.1.3 ¹¹⁹Sb ($T_{1/2} = 38$ h)

Due to the lifetime the samples should be implanted, and taken for measurements and annealing at ISOLDE. Recent technical additions, at the solid state laboratory at ISOLDE, make this a favorable way of doing the measurements. Chambers for making measurements at low temperatures exist in Aarhus and will be transported to ISOLDE, and either the on-line chamber or a new chamber will be used for measurements at temperatures above room temperature. We do not consider us a major user of such a beam, but if it is provided, we would like to have half shift for testing and obtaining data for comparison with other isotopes.

^{119}Sb can be produced with an UC_x target and a Sb RILIS ion source. ^{119}In impurities decay quickly and will not disturb the measurements. About $4 \times 10^8/\mu\text{C}$ for ^{119}Sb are estimated [Koster09].

Another possibility is using a LaC_x target and a MK5 ion source, but this will produce more ^{119}Te , which has a half live of 4.7 days and would therefore hamper studying the Sb behavior in the systems we are interested in [Koster09].

5.1.4 $^{119\text{m}}\text{Sn}$ ($T_{1/2} = 290 \text{ d}$)

Implantations would be performed at ISOLDE and measurements and annealing in home laboratories. As of yet, we have not been requesting this beam for the IS501 experiment, but at this stage it is feasible in order to get data during the 2013 shutdown.

$^{119\text{m}}\text{Sn}$ can be produced with an UC_x target and a Sn RILIS ion source. Yields of ^{119}Sn have been measured $1.8 \times 10^9/\mu\text{C}$ [Koster08]. Both the $^{119\text{g}}\text{Sn}$ and $^{119\text{m}}\text{Sn}$ are produced, but $1.0 \times 10^9/\mu\text{C}$ of $^{119\text{m}}\text{Sn}$ can be expected with some background of ^{119}Cs and ^{119}In , but not at a disturbing level [Koster09].

Molecular beams are not suitable for our measurements.

Production with LaC_x target and Sn RILIS ion source is a possibility with yield of $\sim 1.3 \times 10^7/\mu\text{C}$ of $^{119\text{m}}\text{Sn}$ [Koster09]. This, however, requires much longer implantation time, but would be an option to prepare a weak source if the implantation can be done parallel with other experiments and the UC_x target and Sn RILIS is not available [Koster09].

5.1.5 ^{119}In ($T_{1/2} = 2.1 \text{ min}$)

^{119}In has a very similar half live as ^{57}Mn , and the same setup is used. In 2010 we had UC_x and Ta/W surface ion sources that gave $\sim 4 \times 10^7/\mu\text{C}$, which is on the borderline of what is feasible, and does not allow for TDM's. The laser ionization scheme is fairly simple (one resonance and ionization) [Koster11] and should be possible to set up quickly. Assuming that this gives a factor of 3 compared to W surface ionizer, the yields are estimated to $1.3 \times 10^8/\mu\text{C}$.

5.2 Sample preparation

For the long lived isotopes (^{57}Co and $^{119\text{m}}\text{Sn}$) experience (G. Weyer, private communication) is that a source strength of $\sim 0.1 \text{ MBq}$ ($\sim 3 \mu\text{Ci}$) is the minimum required for having a useful source to use in the laboratory (10-20 measurements of sufficient quality during 1 year).

In the case of $^{119\text{m}}\text{Sn}$ the beam quality at ISOLDE allows us to request a good source strength (0.6 MBq , $\sim 15 \mu\text{Ci}$), with a total dose just below where the transition threshold from amorphous pockets to amorphous layers (around $5 \times 10^{13} \text{ cm}^{-2}$, see e.g. [Weyer82]). With the beam estimates it will be possible to produce two such samples per shift.

For ^{119}Sb , we estimate that for a single implantation, we should have a starting activity of about 370 MBq (10 mCi), which is enough to perform ~ 10 good measurements within ~ 70 hours. This requires $\sim 7.3 \times 10^{12}$ atoms implanted, and is well below the amorphization limit. With the above estimate of $\sim 4 \times 10^8/\mu\text{C}$ $^{119}\text{Sb}/\text{s}$, the making a suitable sample takes of the order of 3-4 hours allowing the preparation of two samples per shift.

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7 Appendix 1: The Mössbauer collaboration at ISOLDE/CERN.

Our team, which initially called itself “The ^{57}Mn Mössbauer collaboration at ISOLDE/CERN” has grown significantly in recent years, and now consist of 14 members directly involved in the Mössbauer measurements. Nine of the members 9 have expertise in running the equipment and the measurements. Hence, we will be able to make use of an increased number of beams, expanding considerably the range and depth of the scientific questions that can be addressed.

The list below contains the members and the most important collaborating partners with respect of the current proposal.

More details can be found on the group web pages (<http://phys.au.dk/forskning/moessbauer-collaboration-at-isoldecern/> [MossbColl]). Note that these pages are incomplete at present, but are expected to be advertised in 2012 on completion.

Country/Institutions	Members	Facilities
Denmark, Department of Physics and astronomy, Aarhus University.	H. P. Gunnlaugsson, G. Weyer	Mössbauer Lab., theoretical calculations of Mössbauer hyperfine parameters (A. Svane).
Iceland, Science Institute, University of Iceland.	S. Ólafsson, H. P. Gíslason, T. E. Mølholt (PhD student) S. Shayestehaminzadeh (PhD student)	Mössbauer Lab., Sputtering growth of various samples, with various types of characterization techniques.
Italy, CNR-IMM MDM Laboratory, Via C. Olivetti 2, 20041 Agrate Brianza (MB)	R. Manotvan	Mössbauer Lab. Growth of samples (ALD) and various characterization techniques. EPR specialists.
South Africa, School of Physics, University of the Witwatersrand; University of KwaZulu-Natal, Durban; iThemba LABS, Somerset West.	D. Naidoo, K. Bharuth-Ram, M. Moodley, H. Masenda (PhD student), M. Ncube (PhD student)	Mössbauer Labs., Ion beam analysis and Magnetization measurement facilities: Specialists in Hyperfine Interactions and Nuclear Methods in Solid State Physics.
Belgium, Instituut voor Kern-en Stralings fysika, University of Leuven.	G. Langouche	Mössbauer Lab., Specialist in Mössbauer spectroscopy (GL).
Switzerland, EP Division, CERN	K. Johnston	Radioactive Laboratory, equipment for annealing and off-line Mössbauer measurements.
Portugal, Departamento Física, Instituto	J. G. Correia (PAC) U. Wahl (EC)	Contact persons regarding PAC and EC experiments

Tecnológico e Nuclear (ITN), Sacavém; IFIMUP, Porto; Centro de Física Nuclear da Universidade de Lisboa (CFNUL), Lisboa	M. B. Madsen	and experiments overlapping with the IS481 experiment physics.
Denmark, Niels Bohr Institute, University of Copenhagen, Copenhagen	M. B. Madsen	Mössbauer Lab., Specialist on relaxation phenomena
France, IM2NP, Aix- , Marseille University	A. Mesli	Specialist on metal defects in silicon

8 Appendix 2: Beam reports 2010&2011

In the following, some relevant information from beam reports from the IS501 experiment are summarized. As the request for opportunistic science may be unorthodox, we have included a description of how that was used and the main outcome.

8.1 2010:

- ^{57}Co ($T_{1/2} = 271$ d): Very high impurity level of ^{57}Fe (~99% of the beam). Limits considerably the application range for this beam for our purposes. Still one sample prepared where the impurity level was beneficial for the interpretations
- ^{57}Mn ($T_{1/2} = 1.5$ min): Very successful, with intensity $\sim 2 \times 10^8$ ions/s. Used up the shifts from the IS443 experiment and started on the new IS501 experiment. Implantation energy had to be limited to 40 keV, which proved useful. Some strange results obtained during this beam-time were repeated in 2011 (50 keV beam) and were not reproducible, meaning that we maintain the wish for ≥ 50 keV beam.
- ^{119}In , no. 1 ($T_{1/2} = 2.1$ min): Failed due to target failure. We were able to demonstrate that our detector system worked.
- ^{119}In , no. 2 ($T_{1/2} = 2.1$ min): Unexpected low intensity ($\sim 2 \times 10^7$ ions/s) limiting the usefulness of this beam. Surprising in view of intensities $\sim 10^9$ ions/s in the 80's [Weyer80-Weyer82]. Still good data acquired, but some experiments impossible. RILIS ionization requested in this addendum.

8.2 2011:

- ^{57}Co ($T_{1/2} = 271$ d): Low intensities and few very weak sources prepared. Still high impurity level of ^{57}Fe (~99% of the beam).
- ^{57}Mn ($T_{1/2} = 1.5$ min): Unusually low intensity $\sim 7 \times 10^7$ ions/s. Extension of the beam time helped in getting our data of high importance. Some data shows unexpected dose dependence, and this is probably due to unusually small beam-spot due to unusually well focused beam [Johnston11]. Clearly we need to make a short implantation of longer lived isotope (e.g. ^{56}Mn) in order to document the size and distribution of the beam-spot. Low intensity of the beam partly due to sharing of the beam with HRS experiment. For the Mössbauer part of ^{57}Mn this is not feasible and results in compromised beamtimes. Optimal conditions for production of ^{57}Mn are with as close to 2 μA of protons as possible.
- ^{119}In ($T_{1/2} = 2.1$ min): Only one measurement performed, intensity estimated to be $< 10^7$ ions/s.

8.3 Beam use

Going through experimental plans and beam reports, Table 3 has been constructed

Table 3: Beam use of the Mössbauer collaboration in 2010 and 2011

<i>Experimental theme</i>	<i>% of beam usage</i>
Paramagnetic relaxations in compound semiconductors	45%

Vacancy diffusion in group IV semiconductors	45%
Calibration	5%
Opportunistic science	5%

The low intensity of the $^{57}\text{Mn}/^{119}\text{In}$ beams in 2011 meant that almost no opportunistic science was performed. As only very minor changes were made to the experimental setup, calibration tasks were simple. With a new detector for ^{57}Fe in 2012, some additional calibration has to be performed. The main opportunistic science 2010-2011 is outlined in Table 4.

Table 4: Main opportunistic science in 2010 and 2011

<i>Task</i>	<i>Hours used</i>	<i>Main findings/outlook</i>
Fe/V superlattices	2	Implantation damage does not destroy the superlattice. Publication being prepared [Mølholt et al.]. Could result in a proposal/Letter of intent after 2013.
III-V nitride semiconductors (InN, AlN)	1	Helps in interpretation of data from GaN, so only partly opportunistic science. Should result in publication [Masenda et al.]
Phase change mechanisms in chalcogenides	1.5	Test performed with ^{57}Mn (^{119}In scheduled for 2011 and cancelled). Major changes during phase change observed, inconsistent with some of the literature. Data being analyzed, and could result in a proposal after 2013 [Mantovan et al.,]
^{57}Mn for Emission channelling	3	Test of ^{57}Mn for Emission channeling experiments. Despite high electron energies, good results were obtained, illustrating the usefulness of this isotope

Other opportunistic science tasks that were on our plan in 2011 were cancelled and the beam time used for obtaining the data for the proposal. This included temperature mapping in Ar implanted ZnO [Bharuth-Ram et al.], search for Fe-Fe dimer in Si [Langouche et al.] and investigation of functional oxide materials [Gunnlaugsson et al.]

8.4 Publications since 2009

Below is a list of reviewed publications since 2009. More details can be found on [MossColl].

1. H. P. Gunnlaugsson, K. Johnston, T. E. Mølholt, G. Weyer, R. Mantovan, H. Masenda, D. Naidoo, S. Ólafsson, K. Bharuth-Ram, H. P. Gíslason, G. Langouche, M. B. Madsen and the ISOLDE Collaboration, Lattice locations and properties of Fe in Co/Fe co-implanted ZnO, *Appl. Phys. Lett.* (2012) Submitted
2. T. E Mølholt, H. P. Gunnlaugsson, K Johnston, R. Mantovan, H. Masenda, D. Naidoo, S. Ólafsson, K. Bharuth-Ram, H. P. Gíslason, G. Langouche, R. Sielemann, G. Weyer and the ISOLDE Collaboration, Spin-lattice relaxations of paramagnetic Fe^{3+} in ZnO, *Physica Scripta* (2012) Accepted.

3. H. Masenda, K. Bharuth-Ram, D. Naidoo, H. P. Gunnlaugsson, T. E. Mølholt, H. P. Gíslason, K. Johnston, R. Mantovan, R. Sielemann, G. Langouche, S. Ólafsson, G. Weyer, the ISOLDE Collaboration, Mössbauer study of ^{119}Sn in $^{119}\text{In}^*$ implanted 3C-SiC, *Hyp. Int.*, (2011) DOI 10.1007/s10751-011-0438-x.
4. K. Bharuth-Ram, W.B. Dlamini, H. Masenda, D. Naidoo, H.P. Gunnlaugsson, G. Weyer, R. Mantovan, T.E. Mølholt, R. Sielemann, S. Ólafsson, G. Langouche, K. Johnston, the ISOLDE Collaboration, ^{57}Fe Mössbauer studies on $^{57}\text{Mn}^*$ implanted InP and InAs, *Nucl. Instr. Meth. B* (2011) In press.
5. T. E. Mølholt, R. Mantovan, H.P. Gunnlaugsson, D. Naidoo, S. Ólafsson, K. Bharuth-Ram, M. Fanciulli, K. Johnston, Y. Kobayashi, G. Langouche, H. Masenda, R. Sielemann, G. Weyer and H.P. Gíslason, Observation of spin-lattice relaxations of dilute Fe^{3+} in MgO by Mössbauer spectroscopy, *Hyp. Int.* **197** (2010) 89-94
6. H. P. Gunnlaugsson, R. Sielemann, T.E. Mølholt, W.B. Dlamini, K. Johnston, R. Mantovan, H. Masenda, D. Naidoo, W. N. Sibanda, K. Bharuth-Ram, M. Fanciulli, H.P. Gíslason, G. Langouche, S. Ólafsson, G. Weyer and the ISOLDE Collaboration, Magnetism in iron implanted oxides: a status report, *Hyp. Int.* **197** (2010) 43-52
7. H. Masenda, D. Naidoo, K. Bharuth-Ram, H.P. Gunnlaugsson, G. Weyer, W.B. Dlamini, R. Mantovan, R. Sielemann, M. Fanciulli, T.E. Mølholt, S. Ólafsson, G. Langouche, K. Johnston and the ISOLDE Collaboration, Mössbauer study of ^{57}Fe in GaAs and GaP following $^{57}\text{Mn}^+$ implantation, *Hyp. Int.* **198** (2010) 15-22
8. H. P. Gunnlaugsson, R. Mantovan, T.E. Mølholt, D. Naidoo, K. Johnston, H. Masenda, K. Bharuth-Ram, G. Langouche, S. Ólafsson, R. Sielemann, G. Weyer, Y. Kobayashi and the ISOLDE Collaboration, Mössbauer spectroscopy of ^{57}Fe in $\alpha\text{-Al}_2\text{O}_3$ following implantation of $^{57}\text{Mn}^*$, *Hyp. Int.* **198** (2010) 5-14
9. H. P. Gunnlaugsson, T. E. Mølholt, R. Mantovan, H. Masenda, D. Naidoo, W. B. Dlamini, R. Sielemann, K. Bharuth-Ram, G. Weyer, K. Johnston, G. Langouche S. Ólafsson, H. P. Gíslason, Y. Kobayashi, Y. Yoshida, M. Fanciulli and the ISOLDE Collaboration, Paramagnetism in Mn/Fe implanted ZnO, *Appl. Phys. Lett.* **97** (2010) 142501, doi:10.1063/1.3490708
10. T. E. Mølholt, R. Mantovan, H. P. Gunnlaugsson, K. Bharuth-Ram, M. Fanciulli, H. P. Gíslason, K. Johnston, Y. Kobayashi, G. Langouche, H. Masenda, D. Naidoo, S. Ólafsson, R. Sielemann, G. Weyer, Temperature and dose dependence of defect complex formation with ion-implanted Mn/Fe in ZnO, *Physica B* **404** (2009) 4820-4822, doi: 10.1016/j.physb.2009.08.187.
11. K. Bharuth-Ram, H. P. Gunnlaugsson, G. Weyer, R. Mantovan, D. Naidoo, R. Sielemann, M. Fanciulli, G. Langouche, S. Olafsson, Th. Aigne, ISOLDE Collaboration, Mössbauer study of Fe in GaAs following $^{57}\text{Mn}^+$ implantation, *Hyp. Int.* **119** (2009) 115–120, doi: 10.1007/s10751-009-9961-4
12. H. P. Gunnlaugsson, G. Weyer, R. Mantovan, D. Naidoo, R. Sielemann, K. Bharuth-Ram, M. Fanciulli, K. Johnston, S. Olafsson, G. Langouche, Isothermal defect annealing in semiconductors investigated by time-delayed Mössbauer spectroscopy: application to ZnO, *Hyperfine Interact.* **188** (2009) 85–89, doi: 10.1007/s10751-008-9893-4
13. D. Naidoo, H. P. Gunnlaugsson, K. Bharuth-Ram, V. V. Naicker, G. Weyer, R. Sielemann, R. Mantovan, M. Fanciulli, the ISOLDE Collaboration, ^{57}Fe Mössbauer

Investigations in P-Type Silicon Germanium Single Crystals. Hyp. Int. **188** (2009)
11-17, doi: 10.1007/s10751-008-9880-9

8.5 Research degrees completed/to be submitted (since 2008):

PhD: R Mantovan, University of Milan, Italy, 2008

D. Naidoo, University of KwaZulu-Natal, South Africa, 2008

T E Molholt, University of Iceland, (to submit in 2012)

MSc: H. Masenda, University of the Witwatersrand, South Africa, 2010

Ms W B Dlamini, University of KwaZulu-Natal, South Africa, 2011.

9 Appendix 3: Description of the proposed experiment

The experimental setup comprises: Mössbauer setup, stored outside the hall when not in use.

Part of the Choose an item.	Availability	Design and manufacturing
Mössbauer setup	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used without any modification
SSP-GLM chamber	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used without any modification <input type="checkbox"/> To be modified
	<input type="checkbox"/> New	<input type="checkbox"/> Standard equipment supplied by a manufacturer <input type="checkbox"/> CERN/collaboration responsible for the design and/or manufacturing

9.1 Hazards generated by the experiment

Additional hazards:

Hazards	Mössbauer setup (online experiments)	SSP-GLM chamber	[Part 3 of the experiment/equipment]
Thermodynamic and fluidic			
Pressure	Low pressure only	Low pressure only	
Vacuum	Yes	Yes	
Temperature	< 100°C (outside setup)	Room temperature	
Heat transfer	No	No	
Thermal properties of materials	Metal	N/A	
Cryogenic fluid	N ₂ , 1[Bar], 3 l/h	N/A	
Electrical and electromagnetic			
Electricity	<20 V, < 20 A	None	
Static electricity	None	None	
Magnetic field	< 0.1 T (outside setup)	None	
Batteries	<input type="checkbox"/>		
Capacitors	<input type="checkbox"/>		
Ionizing radiation			
Target material	Diverse	Diverse	
Beam particle type (e, p, ions, etc)	Ions	Ions	
Beam intensity	<10 ⁹ s ⁻¹	<10 ⁹ s ⁻¹	
Beam energy	>50 keV	>50 keV	
Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:	<input type="checkbox"/>		
• Open source	<input type="checkbox"/>		
• Sealed source	<input type="checkbox"/> [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			

• Description	<input type="checkbox"/>		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity	~50 mCi (inside the setup) (185 MBq)	4-10 μ Ci (0.15-0.4 MBq)	
Non-ionizing radiation			
Laser			
UV light	No	No	
Microwaves (300MHz-30 GHz)	No	No	
Radiofrequency (1-300MHz)	No	No	
Chemical			
Toxic	No	No	
Harmful	No	No	
CMR (carcinogens, mutagens and substances toxic to reproduction)	No	No	
Corrosive	No	No	
Irritant	No	No	
Flammable	No	No	
Oxidizing	No	No	
Explosiveness	No	No	
Asphyxiant	No	No	
Dangerous for the environment	No	No	
Mechanical			
Physical impact or mechanical energy (moving parts)	No	No	
Mechanical properties (Sharp, rough, slippery)	No	No	
Vibration	No	No	
Vehicles and Means of Transport	No	No	
Noise			
Frequency	No	No	
Intensity	No	No	
Physical			
Confined spaces	No	No	
High workplaces	No	No	
Access to high workplaces	No	No	
Obstructions in passageways	Just outside GLM	No	
Manual handling	?	?	
Poor ergonomics	?	?	

0.5 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): ~200 W during on-line experiments