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Nuclear Instruments and Methods in Physics Research B 266 (2008) 3661–3666

NIM B
Beam Interactions
with Materials & Atoms

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High precision ^{180}Hf ion implantation using a high-current ion implanter

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Received 19 October 2007; received in revised form 14 May 2008
Available online 27 May 2008

Abstract

The development of accurate mass spectrometry, enabling the identification of all the ions extracted from the ion source and further precise ^{180}Hf isotope implantation, in a high current implanter is described. The spectrometry system uses two signals (x–y graphic), one proportional to the magnetic field (x-axes), taken from the high-voltage potential with an optic fiber system, and the other proportional to the beam current intensity (y-axes), taken from a beam-stop. The ion beam mass register in a mass spectrum of all the elements magnetically analyzed with the same radius and defined by a pair of analyzing slits as a function of their beam intensity is presented. Hence, it is possible to implant ^{180}Hf^{+}, with less than 1% contamination from neighboring isotopes, in order to conduct material characterization studies by Perturbed Angular Correlations. The precision of the low fluence ion implantation has been done by neutron activation analysis.

PACS: 41.85.Ew; 41.75.Ak; 41.85.Qg; 42.60.Jf; 42.79.Ag

Keywords: Precise ^{180}Hf ion implantation; High current ion implanter; Precise mass spectrometry; Hyperfine interactions

1. Introduction

Nuclear physics applications in materials science using the combined methods of hyperfine interactions studies for the observation of the magnetic and electric interactions between nuclear moments of excited states in specific radioactive probe nuclei and the internal fields of the material and ion channeling for the lattice location studies can be excellent and unique tools for basic studies of defects in single crystals [1]. Any technique, however, has limitations. One needs to introduce the radioactive probe atoms in the lattice to be studied. The radioactive isotope ^{181}Hf is one of the most interesting isotopes to be used and ion implantation is the best way to dope the material when one is interested in using these techniques. The drawback relies on the need to have an available infrastructure for the production of the radioactive ions, such as at CERN/ISOLDE [2] or Bonn [3].

A different approach has been developed in Lisbon with the installation of the high current ion implanter (HCII), model 1090, manufactured by Danfysik [4], at ITN in 1992. Specially oriented for surface engineering studies, the ion implantation facility has been optimized to implant the stable isotope ^{180}Hf. Then the ^{181}Hf radioactive probe is obtained by activation through the reaction ^{180}Hf(n,γ)^{181}Hf using the thermal neutron flux of the Portuguese Nuclear Research Reactor.

In this work, details are given related to the optimization of the equipment for producing the highest possible

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doi:10.1016/j.nimb.2008.05.016

flux of a Hf beam with sufficient mass resolution to implant the isotope ^{180}Hf with a variety of fluences from 10^{12} to 10^{17} ions/cm^2, but minimizing the contribution of neighboring isotopes. A new system for precise identification and recording of the elements extracted from the ion source was developed that was not included in the original configuration of the equipment. More recently, Danfysik installed fully computer control implanters with increased mass spectrometry performance, like the one at the University of Surrey [5], in 1997, and in 2006 at the University of Leuvenn [6]. Details are given about the implantation of $^{180}\mathrm{Hf}^{+}$ and $^{180}\mathrm{Hf}^{++}$ and the respective characterization of the samples using NAA (neutron activation analysis) used to prepare the radioactive samples for the hyperfine interactions studies.

2. Experimental

2.1. Mass spectrometry of the implanter

The high current ion implanter, shown in Fig. 1, was equipped with a beam profiler, an analyzing magnet and *x–y* analyzing slits, enabling a beam envelop imaging and coarse mass analyses of the extracted ion beam. This device, located between the exit of the 90° analyzing magnet and a pair of analyzing slits consists of a probe that intersects the beam, giving an image of the beam horizontal and vertical contours and its position relative to the beam line.

As seen in Fig. 2, application of a magnetic field to the ion beam ensures that ions with different masses extracted from the same source spot hit the analyzing slits in different spots. This physical separation, Δy, between two adjacent ion beams with masses M and $M+\Delta M$, in the analyzing slits, due to the analyzing magnet defection, is known as the magnetic dispersion D [7],

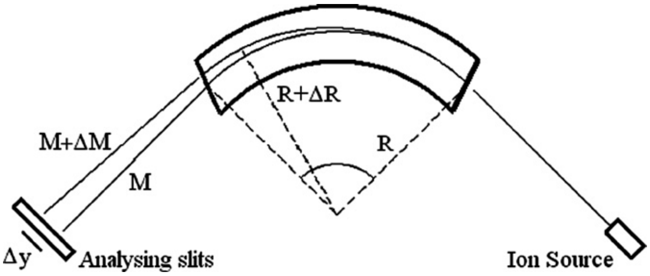

Fig. 2. Schematic of the analyzing magnet ion mass dispersion.

$$D = 2R\frac{M_2 - M_1}{M_1}. \tag{1}$$

where M_1 and M_2 are two adjacent masses (in a.m.u.) and R is the magnet deflection radius.

The dispersion is measured in the analyzing slits, comprising two L-shaped copper bars that are simultaneously moved, having a 0–25 mm variable horizontal aperture and 25 mm constant vertical aperture. It can be seen from Eq. (1) that the dispersion depends only on the magnet geometry and the ion mass.

With the beam profiler is possible to observe the physical separation between the different beam masses, with the possibility of calibration to obtain the mass separation in length units that corresponds to the dispersion value.

The final width, L, of the beam image in the analyzing slits depends on the following factors: (i) extraction power supply stability ($\mathrm{d}E/E$); (ii) magnet current power supply stability ($\mathrm{d}B/B$), (iii) beam divergence α; (iv) initial object width S, and is given by [7]:

$$L = R\frac{\mathrm{d}E}{E} + 2R\frac{\mathrm{d}B}{B} + R\alpha^2 + S \tag{2}$$

where $\mathrm{d}E/E = \mathrm{d}B/B = 5\times10^{-4}$, $R = 500$ mm, $\alpha = 20$ m rad and $S = 4.5$ mm (according to [9] for the extraction

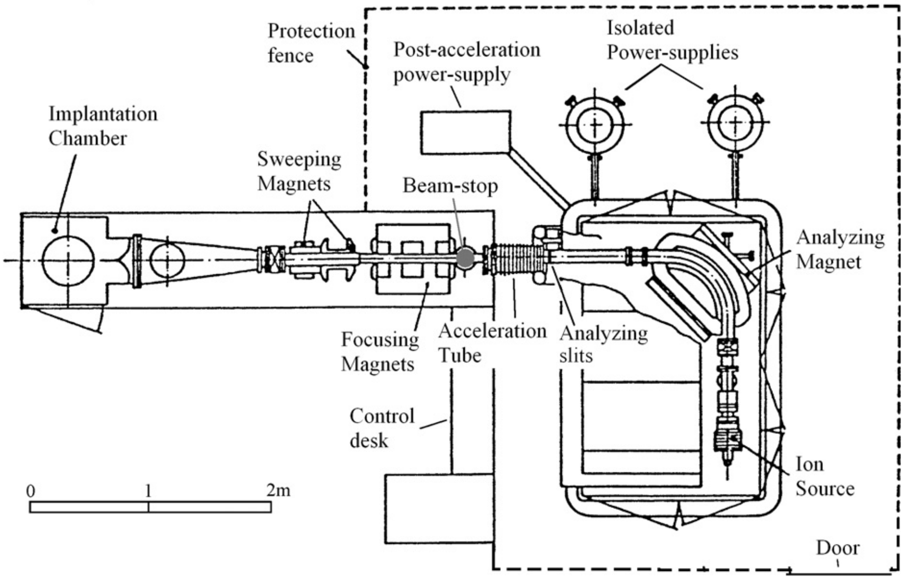

Fig. 1. Layout of the high current ion implanter.

configuration used, the initial object width, S, for a focus beam is about half the extraction electrode aperture), resulting in a beam width of approximately 5.5 mm.

Experimentally, the beam width can change depending on the system operating conditions, where the α and S parameters change in every run. The beam divergence depends on the ion source optimization and the initial extraction aperture width, of 9 mm, which becomes wider and loses its circular shape due to the ion bombardment. All these factors result in an increasing image size and a decrease in the mass separation. Hence, a slit aperture of 7 mm is usually required to transmit about 90% of a mono isotopic beam to the target.

The beam dispersion and width contribute to the implanter mass resolution. The latter is a function of the system geometry and operating conditions, namely the beam focusing. The resolution $M/\Delta M$ can be defined as the mass M for which two adjacent beams of the same current intensity with a image of dimension L in the analyzing slits, and with an atomic mass unit of different ($\Delta M = 1$) can be separated, as shown in Fig. 3.

Two masses can be considered to be resolved if their overlap is less than 10% of the maximum intensity of each (Fig. 3). The resolution can be defined as [7],

$$M/\Delta M = \frac{DM}{L_{90\%}}, \tag{3}$$

where D is the magnetic dispersion (in mm) for mass M (in a.m.u.), and $L_{90\%}$ is the beam width (in mm) (with 90% of the total current density). Considering a beam width of $L_{90\%} = 7$ mm, with the actual operating conditions, the resolution results in approximately $M/\Delta M = 140$, meaning that one can, in principle, separate ^{139}La from ^{140}Ce.

The resolution is fundamental in the operation of an implanter, since it represents the system ability to resolve different masses, allowing for the beam current maximization of each. Thus, if an element maximum beam current intensity cannot be optimized due to some overlap with another element, the implantation is not optimized. Mass resolution has even more relevance in the implantation of heavy ion isotopes. In this case, the problem is the less dispersion (physical separation) between masses.

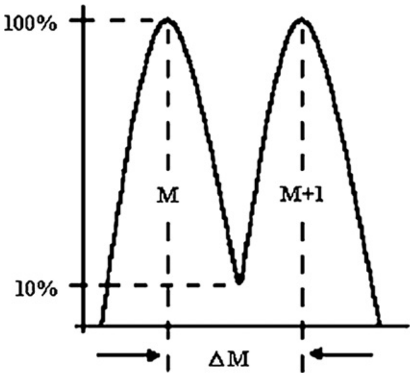

Fig. 3. Schematic for the system resolution, the horizontal beam density was considered as parabolic for simplicity.

However, the beam profiler acting alone does not allow for a global perspective of all the masses that comprise the beam extracted from the ion source. Additionally, since it is located before the analysing slits, any change in the slits aperture does not affect the image of the beam, so there is no direct indication of the mass that strikes the target.

2.2. New mass spectrometry developments

The global record of all the extracted ion species from the source (i.e. a mass spectrum) requires two signals, one proportional to the analyzing magnet magnetic field (x-axes) and another proportional to the beam current intensity (y-axes).

The first signal is taken from a voltage proportional to the analyzing magnet, available from the magnet power supply. The use of this signal comprises two difficulties: (i) the magnet current has hysteresis, thus it is not advisable to reverse the current sense during a mass spectrum; (ii) the analyzing magnet is located on the post-acceleration potential. Therefore, an optic fiber-based circuit was developed to send the magnet current signal to the ground potential. Whereas, on the post-acceleration potential, the d.c. voltage proportional to the magnet current is converted through a voltage-to-frequency (V/F), in a number of voltage pulses with a frequency proportional to its amplitude. This signal is applied to an infrared LED connected to one side of an optic fiber. On the other side, placed at ground potential, a photo-transistor converts the light pulses to a number of voltage pulses with the original frequency. This signal is finally converted in a frequency-to-voltage (F/V) d.c. voltage with the same initial amplitude.

The current signal is taken from a beam stop located just after the acceleration tube, Fig. 1, and not from the non-intersecting device usually used to measure the beam current [8]. The beam stop has no electron suppression but it gives a high accuracy fast electrical signal, considering that the current amplitude is not critical.

3. Measurements

Fig. 4 shows a typical extracted beam mass spectrum when a beam of Hf^{+} is obtained. This spectrum gives a quantitative idea of the main ion species when ions from a high melting point material are obtained using the sputtering technique in the CHORDIS sputter version [9]. These specimens can be divided in four categories: (i) contaminants from the residual air, about 0.5%; (ii) gas used as an auxiliary element to the sputter technique and ionization, in this case Ar (Ar^{+} and Ar^{++}), 95%; (iii) structural species of the source, mainly steel (normally, the most abundant elements are Cr^{+}, Mn^{+} and Fe^{+}), about 1.5%; (iv) sputter target elements, in this case Hf (Hf^{+} and Hf^{++}), about 3%.

For accurate element identification, the calibration of the horizontal scale is done using known reference elements. Normally, the elements used for calibration are:

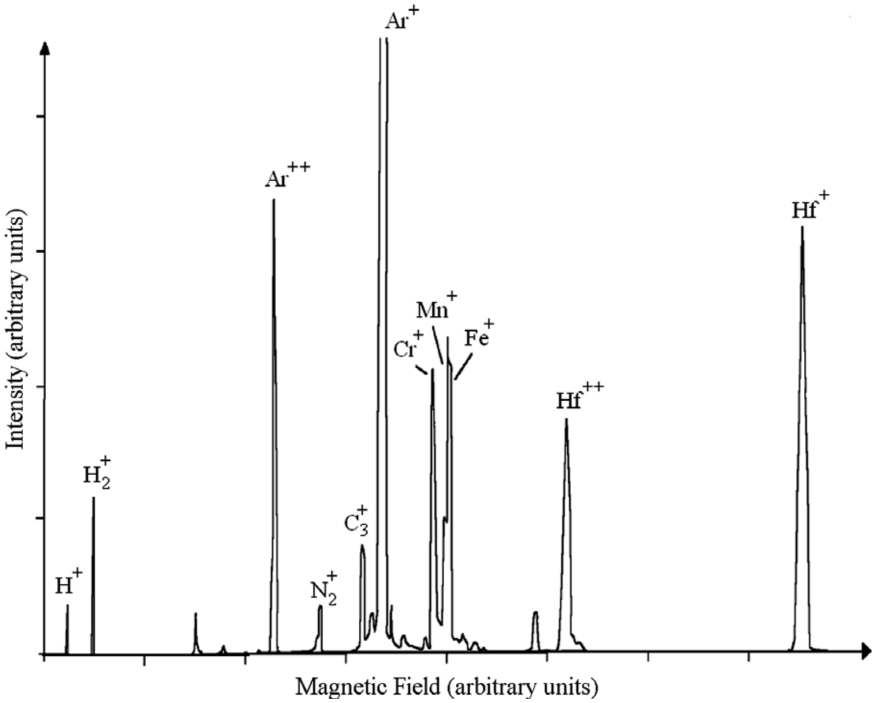

Fig. 4. Total mass spectrum, of the ion beam extracted from the source, running with Hf and Ar, 30 kV extraction voltage and 20 mm slits aperture.

(i) Hydrogen (H_1^+ and H_2^+), the first deflected element by the analyzing magnet and (ii) the gas used in the ion source, usually Ar (Ar^+ and Ar^{++}), which is normally the most intense ion current in the beam. Considering the horizontal axes as a reference, the different line points are given by

$$\text{Calibration} = \sqrt{\frac{M}{q}}, \tag{4}$$

where M is the isotope mass (in a.m.u), q is its charge and the calibration units are arbitrary.

During analysis of a mass spectrum the following should be taken into account: (i) the ion charge state and if it is a molecule; (ii) the magnetic field dependence on $\sqrt{E}$, which results in mass distance change with ion energy, so if the extraction energy if lower, the elements are closer; (iii) that element peak lines are wider for heavier elements. Considering a constant value for $\mathrm{d}B/B$, then for increasing magnetic fields B (i.e. heavier elements) the magnetic dispersion $\mathrm{d}B$ increases, increasing the beam width; (iv) the slits aperture should be smaller than the dispersion to avoid overlapping between adjacent masses that leads to the broadening of the element peak lines in the spectrum.

Considering Fig. 4, the analyzing slits has a 20 mm aperture. Hence, masses below 40 and 41 are resolved, since the dispersion D between mass 40 and 41 is 25 mm. However, isotopes of heavier elements are not resolved, such as Hf which appears as a single peak.

Fig. 5 shows the beam spectrum, with the different Hf isotopes resolved, done with 1 mm aperture on the analyzing slits. Hence, the different Hf isotopes are perfectly resolved, considering that the dispersion for the Hf isotopes is about 5.6 mm. Calculating the Hf isotope abundance from the Hf^+ peak intensities in the spectrum it gives values similar to the natural abundances, with less than 1% error. This indicates that the contamination from other element specimens is low.

For the ^{180}Hf implantation, it is important to know the relative contamination from the other isotopes, as well as the usable slits aperture. The Hf^+ isotopes have an average dispersion of 5.6 mm and a beam width of 7 mm, then the overlapping of the Hf^+ isotopes is inevitable since, as described above, with the actual operating condition the system can separate mass 139 and 140. Consequently, for the precise $^{180}Hf^+$ implantation the slits aperture should be less than 4 mm, with the centre aligned with the $^{180}Hf^+$ isotope peak.

Fig. 5 shows that adjacent to the Hf^+ isotopes, for higher magnetic fields, are some less intense peaks. These correspond to the W^+ isotopes, since W is used in the source filaments. Additionally, as the W filaments are fixed with Ta wires, it is possible to have contamination from $^{181}Ta^+$ in the $^{180}Hf^+$, which is not observed in the mass spectrum of Fig. 5.

4. Results and discussion

Considering that we use the ^{180}Hf implantation for hyperfine interactions measurements, and therefore one needs to activate the sample in a high flux of neutrons in the reactor, it is important that the ^{181}Ta contamination, if exists, is reduced to a minimum. In fact the introduction of ^{182}Ta through the reaction $^{181}\mathrm{Ta(n,\gamma)}^{182}\mathrm{Ta}$ would disturb the measurements.

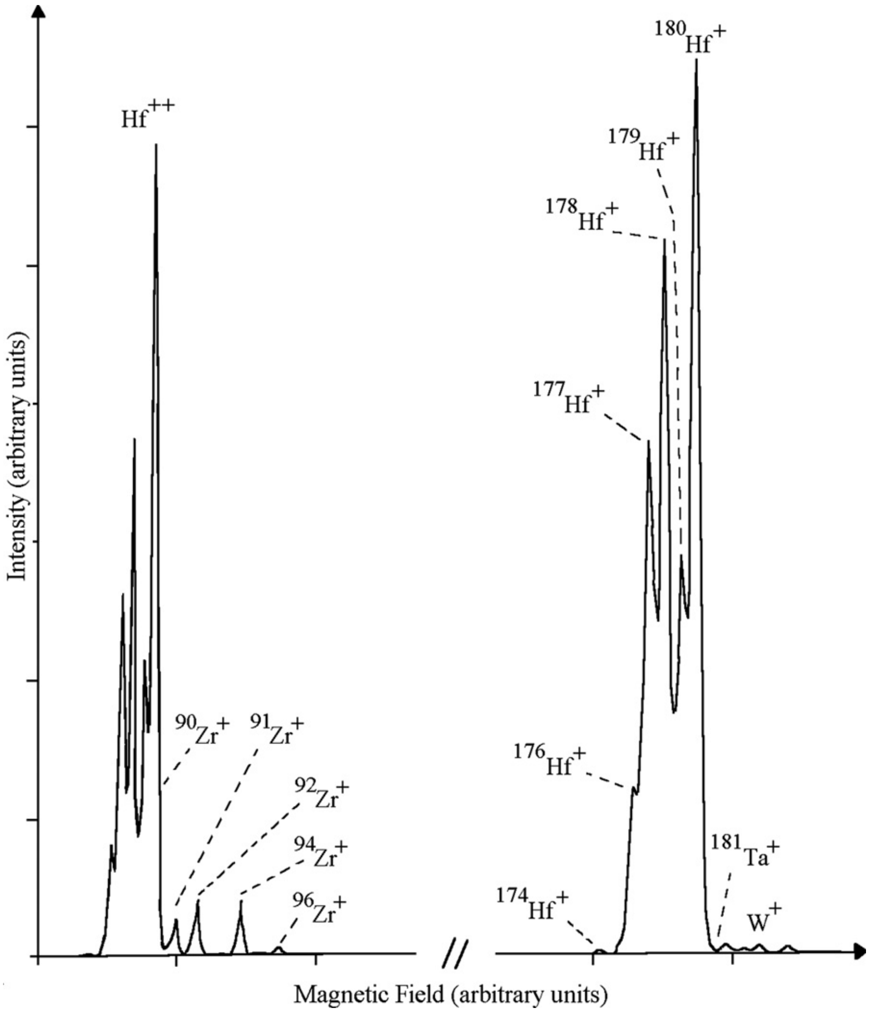

Fig. 5. Hf^+ and Hf^{++} mass spectrum, with 25 kV extraction and 1 mm slits aperture.

For this reason a detailed study of the ^{181}Ta contamination when ^{180}Hf is implanted has been done. First, $^{180}Hf^+$ was implanted in Si samples (fluences from 10^{12} to 10^{17} atoms/cm^2), which were analyzed by NAA (neutron activation analysis). Fig. 6 shows a γ-ray spectrum from one of these samples implanted with $^{180}Hf^+$. The γ-ray energies shown in Fig. 6 spectrum are characteristic of the excited states of ^{181}Ta from the β^- decay of the radioactive ^{181}Hf state. No γ-ray energies of ^{182}Ta: 229.32 keV, 1189 keV and 1122 keV [10] are seen in the spectrum of Fig. 6.

If higher energy is needed during implantation, a Hf^{++} beam should be used. The mass dispersion is the same in Hf^+ and Hf^{++}, as it depends only in the ion mass. Considering Fig. 5, the Hf^{++} peaks are closer, as can be seen from Eq. (1). However, the Hf^{++} beam is contaminated with Zr^+, a natural contaminant of Hf, as can be seen from Fig. 5. This limits the use of the Hf^{++} beam, particularly, the mass $^{180}Hf^{++}$ and $^{90}Zr^+$ (the most abundant Zr isotope), which are deflected with the same radius. From the Zr peak intensities, shown in Fig. 5, and their natural abundance, the $^{180}Hf^{++}$ has 20% of $^{90}Zr^+$, which sets some restrictions to its use.

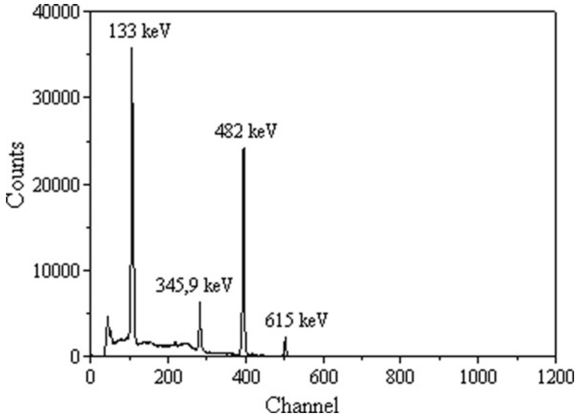

Fig. 6. Implanted $^{180}Hf^+$, 10^{16} atoms/cm^2, Si sample γ-ray spectrum, after neutron activation.

5. Conclusions

The development of an accurate mass spectrometry system that enables the full ion source beam extracted ion species registration (i.e. a mass spectrometry) in a high current ion implanter has been described. The system allows the precise implantation of $^{180}Hf^+$ for hyperfine interactions experiments. This system together with a pair analyzing slits and a beam profiler operation, allows for further increase of implantation accuracy.

Since its installation, we are able to record mass spectra from all the implanted elements and accurate implant specific isotopes. The normal procedure to implant $^{180}Hf^+$ with less that 1% contamination from other ion specimens has been demonstrated. The precision of the ion implantation was determined by NAA.

Acknowledgement

The authors would like to thank António Melo for the fruitful discussions that lead to the understanding of the high current ion implanter operation, and to the previous and today's director of the Ion Beam Laboratory, at ITN, respectively, Maria Fernanda da Silva and Eduardo Alves, for supporting the implanter development and research.

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