

Implantation of the Radioisotope Probe $^{111}\text{In}/\text{Cd}$ as molecular $^{111}\text{InO}^-$ Ions for Materials Studies

Heiko Timmers^{a,*}, Santosh K Shrestha^a, Aidan P Byrne^{b,c}, Wayne D Hutchison^a,
Don H Chaplin^a, Rakesh Dogra^{b,d}

^aSchool of Physical, Environmental and Mathematical Sciences, University of New South Wales at the Australian Defence Force Academy, Canberra, ACT 2600, Australia

^bDepartment of Nuclear Physics, Research School of Physical Sciences and Engineering, Australian National University, ACT 0200, Australia

^cDepartment of Physics, The Faculties, Australian National University, ACT 0200, Australia

^dDepartment of Electronic Materials Engineering, Research School of Physical Sciences and Engineering, Australian National University, ACT 0200, Australia

A low energy (50-150 keV) negative ion implanter at the University of New South Wales in Canberra has been further developed for radioisotope implantations. The new facility has been successfully used to implant the radioisotope probe $^{111}\text{In}/\text{Cd}$ into different material hosts. The probe has been introduced as molecular $^{111}\text{InO}^-$ ions, since the implantation efficiency is three orders of magnitude better than that for elemental $^{111}\text{In}^-$ ion. An implantation rate of $\sim 3 \times 10^4$ Bq/h has been demonstrated. First studies with this radioisotope implanter on bulk silver and indium nitride thin films are reported.

Introduction

Nuclear condensed matter methods such as Perturbed Angular Correlation (PAC), Low Temperature Nuclear Orientation (LTNO) and Nuclear Magnetic Resonance on Oriented Nuclei (NMRO) are well established for the study of electrical, magnetic properties and defect structure of materials [1]. These techniques require a radioisotope probe to be introduced into a host material. The directional distribution of the radiation emitted by the probes can then be detected which can provide information about local electromagnetic fields in the immediate vicinity of the probe.

Radioisotopes are commonly introduced into host materials by thermal diffusion, melting or in-situ irradiation. These methods are not possible for many advanced materials. Moreover, with these techniques there is a limited control on the depth and the number of the probes in the host material. The controlled ion implantation of a radioisotope probe at low energy (< 1 MeV) has several advantages, including reduced sample damage, accurate dosimetry of the number of probing nuclei, and a high degree of control over the implantation depth.

From among the many materials of

current interest which can be studied with radioisotope implantation, indium nitride (InN) may be discussed as an example. InN thin films are topical because of their technological potential [2]. The observed n-type characteristic of as-grown InN is widely attributed to nitrogen vacancies. It has been suggested that InN may contain indium metal clusters, which may obscure the measurement of the electronic band gap [3]. The existence of such clusters can be investigated with PAC at atomic scale. Ion implantation processing may be required for future InN-based devices, as is the case for devices based on Si [4] and GaN [5]. The implantation-induced lattice defects and the annealing behaviour of InN during such processing can also be studied with PAC with $^{111}\text{In}/\text{Cd}$ being an ideal probe.

While positive ion implantation of radioisotopes has been used for materials studies, a cesium-sputtering, negative ion source offers greater versatility since a diverse range of ions can be produced [6-8]. Moreover, with this type of source ion flux can be well controlled which is often not possible with positive ion sources. First results obtained following the negative ion implantation of the probe $^{111}\text{In}/\text{Cd}$ with such a source are reported here.

* Corresponding author. E-mail: H.Timmers@adfa.edu.au

Description of the facility

The radioisotope implanter employs the versatile SNICS-II ion source [9]. Details of the facility are given elsewhere [10,11]. Negative ions are produced in the ion source. They are accelerated by passing through a potential difference of up to 150 kV. Ions are then mass-selected by a 90° dipole magnet and focussed onto a sample with an electrostatic Einzel lens. The total ion current from the ion source (I_s) and the mass selected ion current (I_a) can be measured with Faraday cups before and after the dipole magnet. The implantation of radioisotope ions can be monitored by detecting γ -rays with a NaI(Tl) detector installed outside the target chamber.

The source output of the implanter has been studied in detail for different stable isotopes. An output of 130 μ A has, for example, been achieved for $^{28}\text{Si}^-$ ions. This is about 65% of that reported by Middleton [7]. Similar yields have been obtained for other ions. Mass-resolution of better than 1 u and transmission of mass-selected ions to the target of better than 90% has been achieved.

Radioisotope Implantation

The efficacy of the implanter for the implantation of radioisotopes has been tested with the common PAC probe $^{111}\text{In}/\text{Cd}$. Sustained implantation requires that the radioactivity in the ion source cathode is continuously distributed over a few millimeters. In order to prepare such cathodes, commercially available $^{111}\text{InCl}_3$ solution has been used. As carrier material for the radioisotope ^{111}In , different oxides have been investigated. The oxides were chosen, since they supply ample oxygen for the formation of the molecular $^{111}\text{InO}^-$ ion, which is produced more abundantly than $^{111}\text{In}^-$. Experiments have shown that In_2O_3 powder as a carrier material gives optimum results.

Several new cathode designs have been investigated to optimise radioisotope output of the source. With a cathode in which the cathode recess was conical and the volume was reduced by ~95% compared to the original design (for details see [11]), maximum output and an implantation rate of 3×10^4 Bq/h has been achieved.

In_2O_3 powder was pressed into the

cathode recess, leaving ~1.5 mm above the powder surface to facilitate the pipetting of the $^{111}\text{InCl}_3$ solution. The as-delivered solution, typically 500 mm^3 , was reduced to ~50 mm^3 by evaporation. In several steps the solution was then micro-pipetted onto the pressed powder, which absorbs it readily. After loading the solution, the carrier In_2O_3 was heated to dryness. Measurements have confirmed that more than 90% of the delivered activity is loaded into the cathode.

With the activated cathode in place negative ions were extracted from the ion source and typically accelerated to 125 keV. During beam transport optimisation, the apertures of both the object and the image slits were set at 4mm \times 4mm. Typical ion currents were $I_s = 100 \mu\text{A}$ and $I_a = 6 \mu\text{A}$ for $^{115}\text{In}^{16}\text{O}^-$ ions. During radioisotope implantation the two apertures were opened further (5mm \times 5mm) for increased transmission.

Several materials were implanted with $^{111}\text{InO}^-$ ions including Si, Ag, In metal, InP and InN films. Figure 1 shows a HPGe-detector spectrum from an Ag sample. The 171 keV and 245 keV γ -lines, characteristic of $^{111}\text{In}/\text{Cd}$, are from the sample and verify the implantation of the probe. The inset shows NaI(Tl) spectra from an Ag sample recorded in-situ during implantation which indicate the increase in the implanted $^{111}\text{In}/\text{Cd}$ activity with time.

The efficacy of the implantation of the ^{111}In probe as molecular $^{111}\text{InO}^-$ ions has been demonstrated with LTNO, NMRON

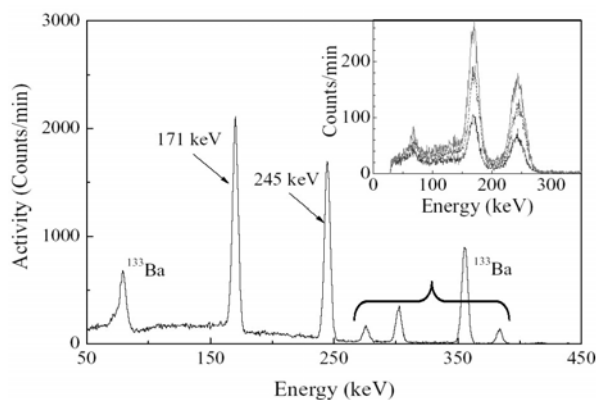


Fig. 1. The partial γ -ray spectrum from a Ag sample implanted with $^{111}\text{InO}^-$ showing the characteristic 171 keV and 245 keV lines. The other lines are from a ^{133}Ba calibration source. The inset shows that count-rate from a Ag sample increases during the $^{111}\text{InO}^-$ implantation.

and PAC measurements. Figure 2(a) shows the results of a brute force LTNO measurement on Ag ($^{111}\text{InAg}$). The anisotropy of the γ -rays is plotted as a function of cool down time of the dilution refrigerator. As expected, a change in the anisotropy is observed for an applied magnetic field B . Increase in B from 6 T to 8 T resulted in an increase of the anisotropy. Comparison with $^{111}\text{InCu}$ data [12] indicates that the implanted ^{111}In nuclei are well thermalised and therefore sited in the host in metallic form.

Figure 2(b) shows the result of a PAC measurement on In metal. The quadrupole interaction frequency of $\nu_Q = 16.6$ MHz derived from the fit of the anisotropy ratio $R(t)$ with theoretical perturbation function agrees with earlier measurements [13].

Local lattice structure of InN films

Figure 3(a) shows $R(t)$ functions for a typical InN film grown by Remote Plasma Enhanced Chemical Vapour Deposition. In this case the implanted fluence of 125 keV $^{111}\text{InO}^-$ radioisotope ions was of the order of 10^{11} ions/cm². However, the total implanted fluence was much larger ($\sim 10^{14}$ ions/cm²) and was dominated by parasitic ions corresponding to neighbouring ion masses. Although a mass resolution of better than 1 u was maintained, tailing of the strong stable ion currents neighbouring the radioisotope into its mass gate could not be prevented. Furthermore after the implantation, discolouration of the film could be observed.

The $R(t)$ measured for this film and shown in Fig. 3(a) can be fitted with $\nu_Q = 17$ MHz corresponding to that of In metal. This indicates that the implanted region of the InN film contains a large amount of metallic In. However, this is inconsistent with the results of Elastic Recoil Detection analysis for these types of films which have shown that pristine quality InN films tend to be highly nitrogen-rich, see [11,14]. The observation is more likely caused by the damage inflicted by the parasitic ions, most likely $^{113}\text{In}^{16}\text{O}^-$ ions. Additional measurements have indeed shown that the observed In signal is due to the depletion of nitrogen from the InN film during the course of the implantation [15].

Following annealing additional higher

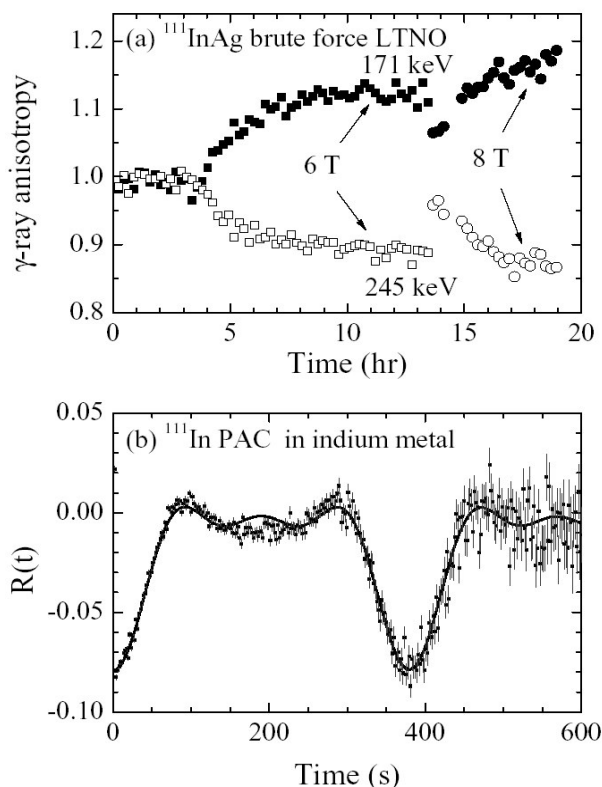


Fig. 2. (a) γ -ray anisotropies from $^{111}\text{InAg}$ brute force LTNO as a function of cool-down time. (b) PAC spectra and fit for the $^{111}\text{In/Cd}$ probe in indium metal. The measurements are consistent with previous results.

frequencies appear (Fig. 3b) which can be best fitted with hyperfine parameters ($\nu_{Q1}=119$ MHz, $\eta_1=0.7$; $\nu_{Q2}=154$ MHz, $\eta_2=0$) corresponding to $^{111}\text{In/Cd}$ on In-sites of an In_2O_3 lattice [16]. In_2O_3 may have formed because of the oxidation of the In metal in the implanted volume. The probe fractions corresponding to the higher interaction frequencies increase with annealing temperature at the expense of that corresponding to In metal.

With additional measurements on a variety of InN films, in which the probe was introduced by $^{111}\text{In}^+$ implantation using the radioisotope implanter at the University of Bonn, Germany and recoil implantation following fusion synthesis at the 14UD accelerator of the Australian National University, it has been determined that the interaction frequency corresponding to the $^{111}\text{In/Cd}$ probe in InN is 28 MHz [17]. A characteristic PAC result from this work is shown in Fig. 3(c).

Present work at the radioisotope implanter of the University of New South

Wales in Canberra concentrates on the development of a new carrier material for the production of $A = 127$ $^{111}\text{InO}^-$ ion, which avoids parasitic background in this mass gate. While aluminium oxide has been shown to be non-suitable, tungsten oxide appears promising.

Conclusions

A radioisotope implantation facility has been developed at the University of New South Wales in Canberra. Successful implantation of the radioisotope probe $^{111}\text{In}/\text{Cd}$ has been demonstrated using negative, molecular $^{111}\text{InO}^-$ ions delivered by a SNICS-II ion source. A suitable design for a sputter cathode has been developed with In_2O_3 powder as the radioisotope carrier. With this cathode design an implantation rate of about 3×10^4 Bq/h has been achieved for $^{111}\text{In}/\text{Cd}$. Measurements have shown that the activity of the $^{111}\text{In}/\text{Cd}$ probe, implanted using the new technique, is suitable for reliable LTNO, NMRON, and PAC.

Implantation into radiation-sensitive semiconductor materials such as InN films resulted, however, in severe damage. This was due to co-implanted parasitic ions tailing into the narrow mass-gate. Radioisotope carrier materials with atomic weights much higher than $A = 127$, for example tungsten oxide, are currently being investigated with the aim to improve implantation conditions.

References

1. G. Schatz, A. Weidinger, Nuclear condensed matter physics: Nuclear methods and applications, John Wiley & Sons, Chichester, 1995.
2. A.G. Bhuiyan, A. Hashimoto, A. Yamamoto, J. Appl. Phys. 94 (2003) 2779.
3. T.V. Shubina et al., Phys. Rev. Lett. 92 (2004) 117407/1.
4. L. Rubin, J. Poate, The Industrial Physicist, June/July (2003) 12.
5. C. Ronning, E.P. Carlson, R.F. Davis, Phys. Rep. 351 (2001) 349.
6. R. Middleton, C.T. Adams, Nucl. Instr. and Meth. 118 (1974) 329.
7. R. Middleton, A Negative Ion Cookbook, University of Pennsylvania, USA (1989).
8. R. Middleton, Nucl. Instr. and Meth. 122 (1974) 35.
9. Instructional Manual: SNICS II Ion Source Model 2JA001110, NEC, Wisconsin, USA (1985).
10. S.K. Shrestha et al., Proceedings of the 13th Australian Conference on Nuclear Techniques of Analysis, ISBN 0-9577217-9-X (26-28 November 2003) Lucas Heights, Australia
11. S.K. Shrestha, PhD thesis, University of New South Wales, Australia (2005).
12. C. Nuytten, Phys. Rev. Lett. 49 (1982) 347.
13. C. Budtz-Jorgensen et al., Phys. Rev. B 8 (1973) 5411.
14. S.K. Shrestha et al., Nucl. Instr. and Meth. B 234 (2005) 291.
15. H. Timmers, S.K. Shrestha, A.P. Byrne, J. Cryst. Growth 269 (2004) 50.
16. W. Bolse, M. Uhrmacher, K.P. Lieb, Phys. Rev. B 36 (1987) 1818.
17. R. Dogra et al., J. Phys.: Condens. Matter 17 (2005) 6037.

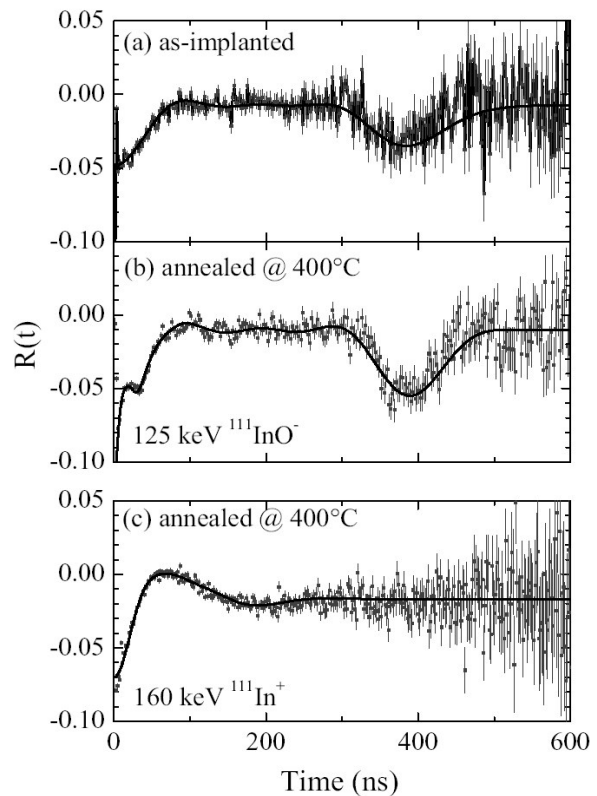


Fig. 3. $^{111}\text{In}/\text{Cd}$ PAC for a RPECVD-grown InN film: (a) as-implanted and (b) after annealing at 400°C following implantation of the $^{111}\text{In}/\text{Cd}$ probe as $^{111}\text{InO}^-$ ions; (c) $^{111}\text{In}/\text{Cd}$ PAC for a similar RPECVD InN film, following the implantation of the probe as $^{111}\text{In}^+$ ions.

Acknowledgements

The authors are grateful to the technical staff of the workshops. Assistance by David Weisser and Vernon Edge and contributions from Darren Smith are also acknowledged.