

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

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Abstract. A radioisotope ion implanter has been developed using a cesium-sputtering, negative ion source, which offers versatility and sustained operation. Employing the molecular $^{111}\text{In}^{16}\text{O}^-$ ion, μCi activities of the radioisotope probe $^{111}\text{In}/\text{Cd}$ have been implanted into different material hosts. The implanted tracer activity has been shown to be sufficient for LTNO, NMRON and PAC. A new NMRON resonance for $^{111}\text{InAg}$ was observed at 75.08 MHz. In_2O_3 powder performed well as the radioisotope carrier in the ion source, with the ratio of radioisotope and parasitic ion current being typically 4×10^{-4} .

Key Words: ion source, LTNO, NMRON, PAC, radioisotope implantation.

1. Introduction

Nuclear condensed matter techniques such as Perturbed Angular Correlation (PAC) spectroscopy, Low-Temperature Nuclear Orientation (LTNO) and Nuclear Magnetic Resonance on Oriented Nuclei (NMRON) require radioisotope tracers to be introduced into a material host as probing nuclei. Though radioisotopes can be diffused into a material, or recoil-implanted following fusion synthesis, the controlled ion implantation of a radioisotope probe has several advantages. They include generally minimal sample damage due to the low implantation energy (50–200 keV), accurate dosimetry of the number of probing nuclei, and a high degree of control over the implantation depth.

For the low energy ion implantation of radioisotope probes, positive ion sources are routinely employed. Since a cesium-sputtering, negative ion source

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[1, 2] offers greater versatility and extended operation, a 50–155 keV ion implanter based on such a source [3] has been equipped and commissioned for radioisotope implantation. While cesium-sputtering ion sources can generally provide large ion outputs, it is not clear, how significant outputs are best achieved for a standard radioisotope probe such as $^{111}\text{In}/\text{Cd}$. Middleton e.g., reports an output of 10 μA of molecular stable $^{115}\text{In}^{16}\text{O}^-$ ions using indium oxide (In_2O_3) as material for the sputter cathode, whereas the best output of elemental $^{115}\text{In}^-$ ions was only 700 nA and obtained using indium metal [2]. Due to the often short half-life of the radioisotope probe, e.g., $t_{1/2} = 2.81$ d for $^{111}\text{In}/\text{Cd}$, and the minute quantities involved, different sputter cathodes are required than those typically used for the production of stable ion beams [2]. Suitable cathode designs and materials have not been reported previously.

In this work the use of a cesium-sputtering, negative ion source for the implantation of the radioisotope probe $^{111}\text{In}/\text{Cd}$ has been studied. The efficacy of this approach for nuclear condensed matter physics has been tested using LTNO, NMRON and PAC.

2. Production and implantation of $^{111}\text{In}^-$ and $^{111}\text{In}^{16}\text{O}^-$ ions

Cesium-sputtering ion sources have been described and studied by Middleton [1, 2]. In such a source Cs^+ ions strike a cooled cathode and sputter material which partially forms negative ions by accepting electrons from cesium atoms. In our experiments both copper and aluminium cylindrical cathode holders (length = 20.8 mm, $\varnothing = 8$ mm) have been tried inside a SNICS-II ion source [3]. The holders have a 7 mm deep recess with an inner diameter of 4.7 mm, which from a depth of 3 mm onwards narrows conically. This matches the sputter crater of the Cs^+ ions.

As cathode material and carrier of the ^{111}In radioisotope, powders of Al, Al_2O_3 and In_2O_3 have been investigated. Compressed Al powder, even with admixtures of Al_2O_3 , is not sufficiently absorbent to function as carrier. While it avoids the stable indium isotopes $^{113,115}\text{In}$ and their molecular ions, the slow sputter-rate of Al_2O_3 and the extremely high $^{16}\text{O}^-$ output has proven this also a bad choice. The use of In_2O_3 as cathode material has produced the best results. Figure 1(a) shows a mass spectrum for this cathode. The relative yields of the two molecular ions $^{113}\text{In}^{16}\text{O}^-$ and $^{115}\text{In}^{16}\text{O}^-$ reflect the natural abundance of the two stable indium isotopes. Notably, the yield of the elemental $^{115}\text{In}^-$ ion is three orders of magnitude smaller than that of the molecular ion $^{115}\text{In}^{16}\text{O}^-$. The $^{113}\text{In}^-$ current was not observed.

The In_2O_3 powder was pressed into the cathode holder recess. Approximately 1.5 mm were left above the powder surface to facilitate the loading of the radioisotope solution. Activities of 5 mCi of $^{111}\text{In}/\text{Cd}$ were purchased as $^{111}\text{InCl}_3$ in 0.05 molar HCl ¹. The volume of the solution was reduced to typically 50 mm^3

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using a 150 W heat lamp at a distance of 10 cm. In several steps the solution was then micro-pipetted onto the surface of the pressed In_2O_3 carrier, which absorbs the solution quickly. Experiments with diluted ink and radiographs of used cathodes suggest that the loaded activity is essentially homogeneously distributed throughout the carrier. Following 45 min drying under a 150 W heat lamp, the activated cathode was placed inside the ion source. Measurements have shown that 90% of the activity are loaded into the cathode. The missing fraction remains in the delivery bottle and the pipette.

The ion source was operated with typical parameters [3]. Negative ions were extracted from the source, accelerated to an energy of 125 keV, and focused through a quadratic aperture (4 mm \times 4 mm) into a Faraday cup, with which the source output current I_s was measured. Mass analysis was achieved with a 90° dipole magnet. Individual mass numbers A were selected (Figure 1(a)) using an aperture (3 mm \times 3 mm) after the magnet. The analysed current I_a was measured with a second Faraday cup. Typical beam 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 (5 mm \times 5 mm) for increased transmission.

The field of the magnet was calibrated using $^{16}\text{O}^-$, $^{113}\text{In}^{16}\text{O}^-$, $^{115}\text{In}^{16}\text{O}^-$ and $^{133}\text{Cs}^-$ beams. Both $^{111}\text{In}^-$ and $^{111}\text{In}^{16}\text{O}^-$ ions have been implanted into a variety of materials. As suggested by Figure 1(a) the source output of elemental $^{111}\text{In}^-$ ions was too small to achieve practically useful implanted activities. Hence the $^{111}\text{In}/\text{Cd}$ probe has been implanted as molecular $^{111}\text{In}^{16}\text{O}^-$ ion. Implanted activities were of the order of 1 μCi , with activity increases in the range of 0.02–0.15 $\mu\text{Ci}/\text{h}$, and a maximum implanted activity of 2.3 μCi . Figure 1(b) shows a γ -ray spectrum for this sample. The analysed current I_a for the mass number of $^{111}\text{In}^{16}\text{O}^-$ ($A = 127$) was dominated by parasitic ions, as it is demonstrated in Figure 1(c). The ratio of radioisotope to parasitic current was typically 4×10^{-4} . Implanted fluences, including the parasitic ions, were several 10^{14} ions/cm². In the literature e.g., implanted fluences of the order of 10^{13} ions/cm² are quoted for the low energy implantation of the $^{111}\text{In}/\text{Cd}$ probe for PAC using a positive ion source and $^{111}\text{In}^+$ ions [4]. While $^{111}\text{Cd}^{16}\text{O}^-$ ions are likely to constitute a fraction of the parasitic ion current, a much larger fraction must have an origin unrelated to the $^{111}\text{In}/\text{Cd}$ probe. The identification and suppression of this latter fraction would improve the technique.

3. Radioisotope tracer studies with $^{111}\text{In}/\text{Cd}$

The efficacy of the implantation of $^{111}\text{In}^{16}\text{O}^-$ ions to introduce the $^{111}\text{In}/\text{Cd}$ probe has been demonstrated with LTNO, NMRON and PAC measurements on a variety of samples. Importantly, the co-implantation of a significant number of parasitic ions does generally not interfere with the application of these techniques. Samples of the radiation-sensitive semiconductor indium nitride were, however, severely depleted of nitrogen in the implanted volume [5].

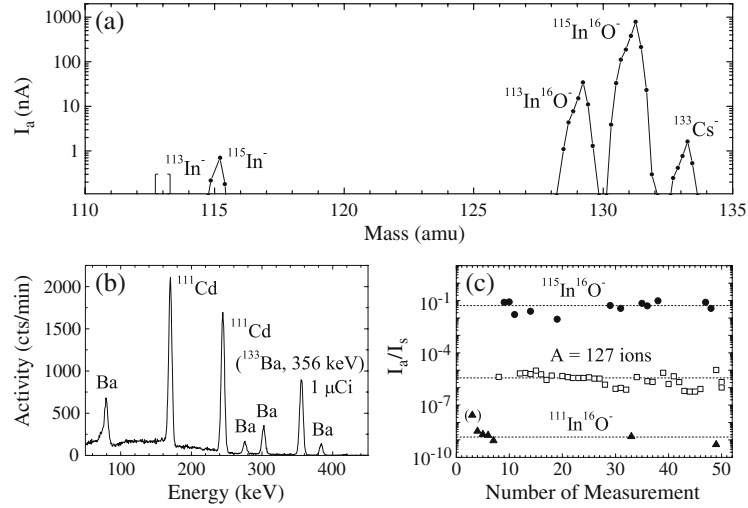


Figure 1. (a) Partial mass spectrum for a Cs-sputtering ion source using an In_2O_3 cathode. (b) γ -ray spectrum from a sample implanted with $^{111}\text{In}^{16}\text{O}^-$ ions demonstrating an implanted activity of $2.3 \mu\text{Ci}$. Additional lines are from a calibrated ^{133}Ba source. (c) The $^{111}\text{In}^{16}\text{O}^-$ current compared to that of the parasitic ions ($A = 127$) and that of $^{115}\text{In}^{16}\text{O}^-$ from many measurements. The $^{111}\text{In}^{16}\text{O}^-$ current has been derived from the implanted activity. Currents have been divided by the source output I_s .

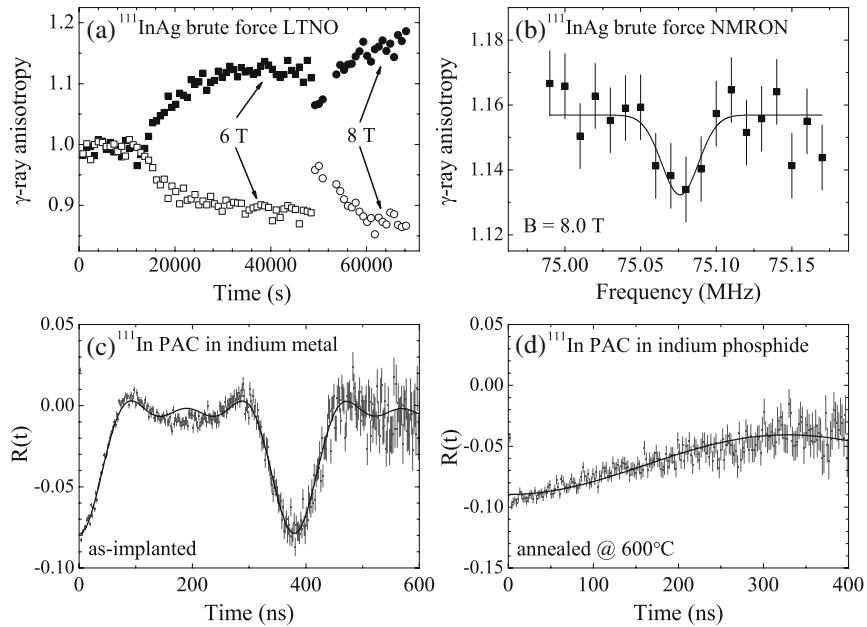


Figure 2. (a) γ -ray anisotropies from $^{111}\text{InAg}$ brute force LNTNO as a function of cool-down time. (b) γ -ray anisotropy from $^{111}\text{InAg}$ brute force NMRON. A gaussian fit indicates a resonance at 75.08 MHz. (c) PAC spectrum and fit for $^{111}\text{In}/\text{Cd}$ in indium as measured at room temperature. (d) PAC spectrum and fit for $^{111}\text{In}/\text{Cd}$ in indium phosphide.

Figure 2(a) shows the results of a brute force LTNO measurement on silver ($^{111}\text{InAg}$) following the implantation of $^{111}\text{InO}^-$ ions to an activity of $0.8 \mu\text{Ci}$. Comparison with $^{111}\text{InCu}$ data [6] indicates that the ^{111}In nuclei are well thermalised and therefore sited in the host in metallic form. Following a second implantation, resulting in a probe activity of $2.3 \mu\text{Ci}$, the first brute force NMRON measurement with $^{111}\text{In}/\text{Cd}$ was performed on silver in a magnetic field of 8.0 T. The anisotropy for the 171 keV γ -ray line of ^{111}Cd is shown in Figure 2(b). Though statistics are poor, the resonance may be identified at 75.08 MHz. This frequency is in agreement with that predicted by the $^{111}\text{InCu}$ slope to three significant figures.

For PAC spectroscopy, among other hosts, indium and indium phosphide were implanted with $^{111}\text{In}^{16}\text{O}^-$ ions. The anisotropy ratios $R(t)$, determined as described in Ref. [7], are shown in Figure 2(c) and (d), respectively. Least-squares-fits with the appropriate theoretical perturbation function are also shown. The observed quadrupole interaction frequency of $\nu_Q = 16.6 \text{ MHz}$ for indium agrees with earlier measurements [8]. As expected, for indium phosphide a cubic environment of the probe nuclei is seen [7, 9].

4. Conclusions

Low energy ion implantation of the radioisotope tracer $^{111}\text{In}/\text{Cd}$ has been demonstrated using molecular, negative $^{111}\text{In}^{16}\text{O}^-$ ions delivered by a cesium-sputtering source. A suitable design for a sputter cathode has been developed with In_2O_3 powder as the radioisotope carrier. The ratio of the $^{111}\text{In}^{16}\text{O}^-$ ion current to parasitic ion current of the same mass was found to be typically 4×10^{-4} for the set-up used. Efforts to reduce the parasitic fraction of the ion flux by employing alternative radioisotope carriers are underway. Measurements for several material hosts have shown that μCi $^{111}\text{In}/\text{Cd}$ activities implanted using this technique are sufficient for reliable LTNO, NMRON and PAC. A new NMRON resonance for $^{111}\text{InAg}$ was observed at 75.08 MHz.

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