Implantation of ^{111}In in the Heusler Alloys Pd_2MnZ (Z=Sn,Sb,Ge,In) Following Heavy Ion Nuclear Reactions: Measurement of Magnetic Hyperfine Field using PAC Spectroscopy

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Ion implantation of the recoil ^{111}In nuclei following heavy ion nuclear reactions ^{108}Pd(^{7}Li,4n)^{111}In and ^{108}Pd(^{6}Li,3n)^{111}In has been used to implant ^{111}In probes in the Heusler alloys Pd_2MnZ(Z=Sn,Sb,Ge,In). Perturbed Angular Correlation method was used to study the hyperfine magnetic field in these alloys. Direct implantation of ^{111}In probe nuclei was used to great advantage in the present case resulting in large implantation efficiency. Only a few hours of irradiation time with moderate beam current of the order of 400-500 nA resulted in sufficient implanted ^{111}In activity on the sample for good quality measurements. The hyperfine field was measured at ^{111}In probe nuclei substituting Mn and Z sites as a function of temperature. The fraction of ^{111}In nuclei occupying Mn atom sites was found to increases with the annealing of sample at higher temperatures.

I. INTRODUCTION

The radioactive probe nuclei, used in the study of hyperfine interactions with Perturbed Angular Correlation (PAC) spectroscopy, are generally produced through nuclear reactions using particle accelerators or nuclear reactors. These radioactive nuclei are then introduced in to the samples to be studied using a variety of chemical and metallurgical processes or through ion implantation. The ion implantation is usually carried out either by using accelerated radioactive ion beams or through radioactive ions recoiling out of the target following heavy ion nuclear reactions.

The ion implantation process is particularly advantageous because it introduces radioactive probes into an already prepared sample, which avoids extensive manipulation of the radioactive material. In the present work direct implantation of ^{111}In nuclei in the samples was achieved through nuclear reaction ^{108}Pd(^{6}Li,3n)^{111}In or ^{108}Pd(^{7}Li,4n)^{111}In, using 8 UD Pelletron Tandem Accelerator at the Institute of Physics of the University of São Paulo.

Besides reporting a new and efficient way to introduce ^{111}In probe nuclei into samples for PAC measurements, we also show that the method of introducing the probe can influence the final site location of the probe and give different results. In order to test the method of implantation we have used Pd-based Heusler alloys as samples and studied the local magnetism by measuring the hyperfine fields with PAC technique.

II. EXPERIMENTAL PROCEDURE

A. Ion Implantation

The technique of recoil-ion-implantation of PAC probes, following heavy-ion nuclear reactions, has been used efficiently to implant the radioactive isotope ^{111}In in semiconductor samples [1]. In this method the authors used a thin foil of Rh as target and bombarded it with a ^{12}C beam of 69 Mev. The short lived radioactive nuclei ^{111}Sn ($T_{1/2}$ = 35 min) and ^{111}Sb($T_{1/2}$ = 1.3 min), produced in nuclear reactions ^{103}Rh(^{12}C, p3n)^{111}Sn and ^{103}Rh(^{12}C, 4n)^{111}Sb having large recoil energy ($\sim$7 MeV), exit thin Rh foil (2-3 micrometers) and get implanted on substrate kept behind the target. These short-lived nuclei eventually decay to the desired probe nuclei ^{111}In ($T_{1/2}$ = 2.8 d). The obvious advantages of these reactions are the high natural abundance of ^{12}C and ^{103}Rh, 98.9 % and 100 %, respectively and relatively high recoil energy of the product nucleus.

The Pelletron Tandem accelerator at the Physics Institute of the University of São Paulo, used in the present work however, can accelerate ^{12}C beam to only 56 MeV, which is not sufficient to produce ^{111}In in good yield. Given the limitation it was decided to try alternate reactions like ^{108}Pd(^{7}Li, 4n)^{111}In and ^{108}Pd(^{6}Li, 3n)^{111}In, using ^{6}Li and ^{7}Li beams with maximum available energy of 32 MeV.

It was also decided to use the natural Pd target having ^{108}Pd with $\sim$27 % of abundance, in the preliminary experiments to implant ^{111}In in a series of Heusler alloys of the type Pd_2MnZ(Z=Sn,Sb,Ge,In). These alloys have a cubic $L2_1$ structure and order ferromagnetically with a magnetic moment of about 4.3 μ_B localized on Mn. The Heusler alloys $Pd_2MnSb(Sn)$ have been investigated in the past with PAC spectroscopy [2, 3]. The radioactive ^{111}In probe, introduced in the samples during its preparation by induction melting of component elements, was found to substitute only the Sn and Sb atom sites. On the other hand when ^{111}Ag was introduced in Pd_2MnSn sample through thermal diffusion it occupied the Mn site [4].

In the present experiment heavy ion nuclear reactions ^{108}Pd(^{7}Li, 4n)^{111}In and ^{108}Pd(^{6}Li,3n)^{111}In, in which Pd_2MnZ(Z=Sn,Sb,Ge,In) Heusler alloys themselves served as the reaction target, was used to implant the recoiling ^{111}In nuclei in to the sample. Calculations made with the program PACE for the fusion-evaporation reaction cross sections shown in Fig. 1 indicated that the integral cross-section for the production of ^{111}In is considerably larger for ^{6}Li beam compared to ^{7}Li at all energies above threshold. The ^{6}Li beam was therefore chosen for all the experiments.

Since the Heusler alloys chosen for the experiment all contain Pd as one of the component elements, they themselves served as reaction targets. The samples were cut in to small slices of about 5x5 mm^2 and 1mm thick and mounted in an especial reaction chamber[5] for irradiation with the ^{6}Li beam. The average recoil energy of ^{111}In ions being too small ($\sim$1.7 MeV) they all stop in the relatively thick ($\sim$1 mm) sample and get directly implanted in the Heusler alloy. This is a great advantage in the present case, as the implantation efficiency tends to be almost 100 %. With a beam current of 400-500 nA only a short irradiation time of the order of 8-10 hours was found sufficient to implant more than 20 μCi of ^{111}In in the samples for a good quality PAC measurement.

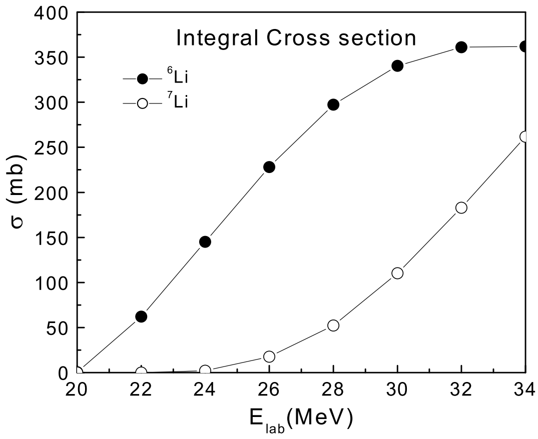

FIG. 1: Cross-sections for the collision of a ^{6}Li and ^{7}Li beams with ^{108}Pd target, as a function of the beam energy.

III. EXPERIMENTAL RESULTS

Since the Heusler alloys Pd_2MnZ(Z=Sn,Sb,Ge,In) used in the present experiment contain, apart from Pd, also other elements such as Mn,Sn,Sb,Ge and In, all of them in their natural isotopic composition, it was realized that nuclear interaction of ^{6}Li beam with these nuclei would produce several other radionuclides apart from ^{111}In. Depending on the half-lives and gamma rays emitted in their decay these radionuclides could seriously interfere with the PAC measurements.

The low energy gamma ray spectra for some of the Heusler alloys taken with a Ge(HP) detector spectrometer, 10-12 hours after the end of irradiation, are shown in Fig. 2. All the spectra show gamma rays at 171 keV and 245 keV belonging to ^{111}In ($T_{1/2}$ = 2.8 d) and a gamma ray at 203 keV belonging to ^{109}In ($T_{1/2}$ = 4.2 h) resulting from nuclear reaction with Pd. The principal gamma rays resulting from nuclear reactions with Sn, Ge and In come from ^{123}I ($T_{1/2}$ = 13 h), ^{77}Br ($T_{1/2}$ = 57 h), ^{73}Se ($T_{1/2}$ = 7 h) and ^{118}Sb($T_{1/2}$ = 5 h) as can be seen in Fig. 2.

Since PAC measurements started about 24-30 hours after the end of irradiation, important conclusion is that none of these additional gamma rays interfered with the PAC experiments since it involved gamma-gamma coincidence measurements of the 171- 245 keV gamma cascade in the decay of ^{111}In.

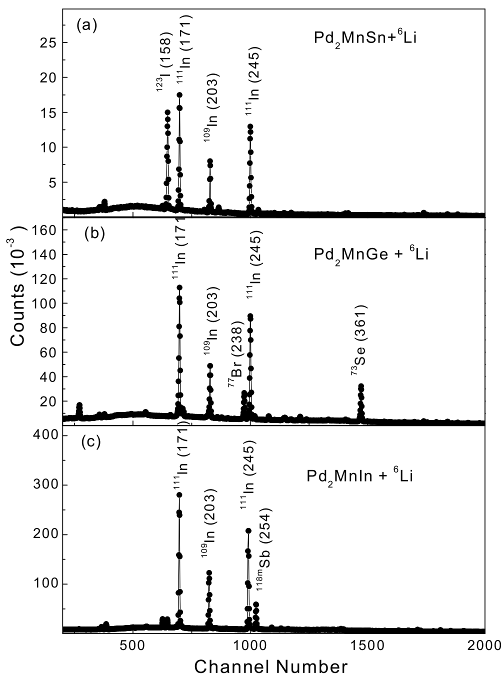

FIG. 2: Gamma ray spectra of the sample of Pd_2MnSn(Ge,In) measured with GeHP detector 10-12 hours after ion implantation.

TDPAC measurements were carried out at the Hyperfine Interaction Laboratory at IPEN using a spectrometer consisting of four BaF_2 detectors and associated electronic set up generating simultaneously 12 delayed coincidence spectra. Details about the PAC measurements can be found else where [6, 7]. The spectrum taken for the as implanted sample of Pd_2MnSn at 295K shown in Fig. 3(a) is a typical one for a radiation damaged sample in which the amplitude of the ratio R(t) $\approx$ $A_{22}G_{22}(t)$, where $G_{22}(t)$ is the perturbation coefficient, shows rapid attenuation. All the irradiated samples were thermally annealed at 400 ^{o}C for 24 hours before starting the PAC measurements to eliminate or substantially reduce the radiation damage effects. The spectrum given in Fig. 3(b) shows an almost complete recovery of the oscillation amplitude after annealing. A slow attenuation still observed in the curve might be the result of a low frequency quadrupole interaction present in the sample due to some disorder or structural defects but does not interfere in the determination of magnetic hyperfine field.

TDPAC spectra for the Heusler alloys

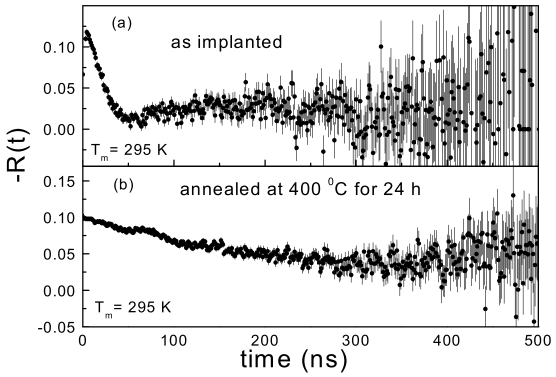

FIG. 3: Measured ratio functions for Pd_2MnSn, as implanted (a) after annealing at 400 ^oC (b)

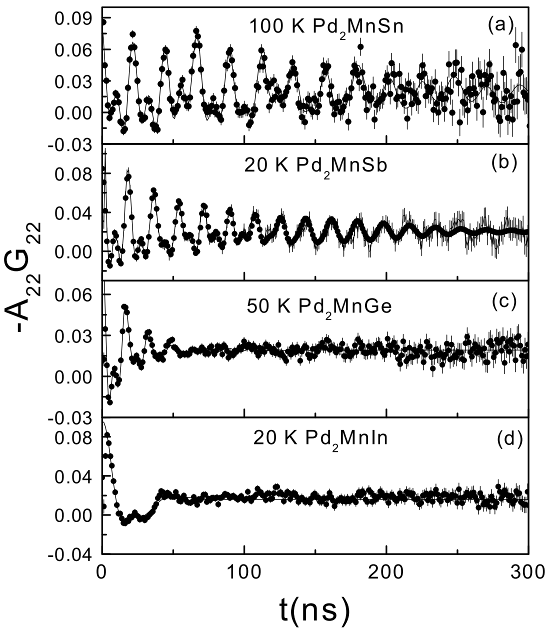

FIG. 4: TDPAC spectra for Heusler alloys after annealing at 400C (a,b,c) and after an additional annealing at 800 ^oC (d)

Pd_2MnZ(Z=Sn,Sb,Ge,In) taken at temperatures below magnetic transition temperatures are shown in Fig. 4. Detailed analysis of these spectra showed two magnetic interactions in the case of alloys Pd_2MnSn(Sb,Ge) which were assigned to ^{111}In probe occupying Mn and Sn(Sb,Ge) sites respectively.

As expected Pd_2MnIn alloy did not show magnetic interaction. This is due to the fact that ^{111}In is substituting some of the In atom in this alloy and the crystal structure of the alloy is such that the In atom is in between the two layers of Mn atoms having opposite spins. Due to opposing spins of Mn atoms there is no net transfer of spin density to the probe resulting in zero hyperfine field at In site. After an additional annealing of the sample at 800 ^oC for 12 h, PAC measurements showed a unique frequency in this alloy, which was assigned to ^{111}In probe nuclei occupying the Mn sites as shown in Fig. 4(d). The migration of radioactive probe from transition element site to Mn site at higher temperature annealing was observed in all the alloys. This was seen from the increase in the fractional occupation of Mn sites relative to transitional element site after higher temperature annealing.

IV. CONCLUSION

Present experiment has demonstrated that for samples where Pd is one of the components, the process of ^{111}In implantation using the present nuclear reaction in thick target is quite efficient compared to conventional methods of introducing the probe nuclei in the sample. The main reason for this is the relatively low recoil energy ($\sim$1.7 MeV) imparted to the reaction product. Most of the ^{111}In recoils therefore stop in the target(sample)itself. On the other hand for example, conventional ion implanter which produces radioactive ^{111}In ion beam for implantation has very small efficiency due to low beam transmission characteristics of these machines (of the order of 0.1%) or less. As a consequence one needs to use very high specific activity of ^{111}In in the ion source. In heavy ion nuclear reaction method, high recoil energies are necessary in order to implant the probe nuclei in substrate kept behind the target. About 50-60% of all nuclei produced in target often get implanted in samples.

For the implantation of ^{111}In on samples that do not contain Pd the method will require some modifications. The ^{108}Pd(^{6}Li, 3n)^{111}In reactions could be produced in a thin foil of Pd (preferably enriched in ^{108}Pd) and swift ^{111}In ions recoiling out of the foil may be stopped in the substrate placed behind the target at a suitable distance and geometry. The reaction chamber for such experiments is under test. Low energy of ^{111}In recoils may pose serious problems however, in terms of the efficiency of the process.

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