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Implantation of ^{111}In-probe Nuclei with Nuclear Reactions ^{108}Pd(^{6,7}Li, xn)^{111}In using Pelletron Tandem Accelerator: Study of Local Magnetism in Heusler Alloys

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Abstract. Perturbed Angular Correlation method has been used to study the hyperfine magnetic field in the Heusler alloy $Pd_2MnSb(Sn)$. 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 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}Cd probe nuclei substituting Mn and Sb(Sn) sites as a function of temperature. The fraction of ^{111}Cd nuclei occupying Mn atom sites was found to increases with the annealing of sample at higher temperatures.

Key Words: Heusler alloys, magnetic hyperfine field, PAC, probe implantation.

1. Introduction

There are several different ways to introduce radioactive probe nuclei into samples to be measured by Perturbed Angular Correlation (PAC) spectroscopy, each one having its own advantages and disadvantages. 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. What is still a subject of investigations is how the method of introducing probes influences its site location. In the present work, 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 give different results. In order to test the method of implantation we have used Pd-

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based Heusler alloy as sample. The Heusler alloys $Pd_2MnSb(Sn)$ have a cubic $L2_1$ structure and order ferromagnetically with a magnetic moment of about 4.3 μB localized on Mn. These alloys have been investigated in the past with PAC spectroscopy [1, 2]. 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 [3]. 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 $Pd_2MnSb(Sn)$ Heusler alloys themselves served as the reaction target, was used to implant the recoiling ^{111}In nuclei in to the sample.

2. Experimental

The Heusler alloys were prepared by arc-melting the stoichiometric amounts of the constituent elements (99.99% purity) in argon atmosphere purified with a hot titanium getterer. After annealing at 800°C during 24 h in the argon atmosphere the samples were analyzed by X-ray powder difraction, and were found to be essentially single phase with the correct $L2_1$ structure. The ^{111}In nuclei were implanted in the samples by means of nuclear reactions $^{108}Pd(^{7}Li, 4n)^{111}In$ or $^{108}Pd(^{6}Li, 3n)^{111}In$, with a beam energy of 32 MeV using eight UD Pelletron Tandem Accelerator at the Physics Institute of the University of São Paulo. Since Pd is a constituent element of the alloys in the present experiment, the samples themselves served as the reaction targets during irradiation and practically all the ^{111}In nuclei produced in the reaction stopped in the sample. The Heusler alloy samples were cut in the form of a 5 mm diameter disc with a thickness of ∼1 mm and mounted in a special reaction chamber [4]. The irradiation times varied from 8–10 h with an average beam current of the order of 400–500 nA on the target. A few hours after the end of irradiation, the samples were examined with HPGe detector for the presence of other radioactive nuclei produced through side reactions with other elements of the sample in addition to the desired ^{111}In. Most of the radioactive products found were either short lived or had little interference in the measurement of coincidence spectra of the gamma–gamma cascade in the decay of ^{111}In.

The TDPAC measurements were carried out with a conventional fast–slow coincidence set-up using four conical BaF_2 detectors. The gamma cascade of 172–245 keV populated from the electron capture decay of ^{111}In, and having an intermediate level at 245 keV with spin $I = 5 / 2^+$ and $T_{1/2} = 85$ ns, in ^{111}Cd was used to measure the magnetic hyperfine field. The samples were measured in the temperature range of 20–295 K using a closed-cycle helium cryogenic device. The time resolution of the system was about 600 ps.

The TDPAC method is based on the observation of hyperfine interaction of nuclear moments with extra-nuclear magnetic field or electric field gradient. A detailed description of the method can be found elsewhere [5, 6]. The per-

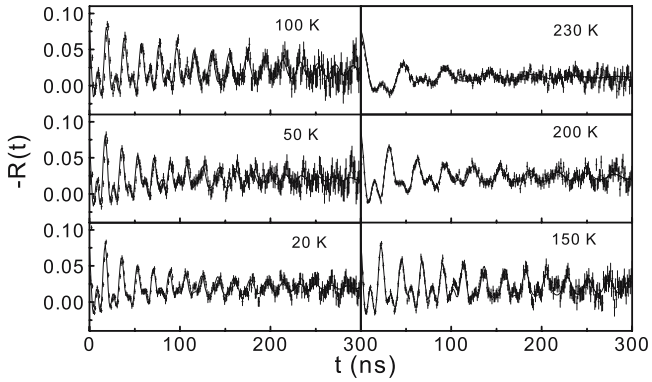

Figure 1. TDPAC spectra of ^{111}Cd implanted in Pd_2MnSb at different temperatures.

turbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. Measurement of $G_{22}(t)$ allows the determination of Larmor frequency $\omega L = \mu_N g B_{hf} / \hbar$. From the known g-factor, $g = 0.306$ of the 245 keV state of ^{111}Cd it is thus possible to determine the magnetic hyperfine field B_{hf}.

3. Results and discussion

Some of the TDPAC spectra measured at different temperatures for the alloy Pd_2MnSb are shown in Figure 1. The samples were annealed at 400°C in vacuum for about 4 h, after implantation, before starting the PAC measurements to eliminate the effects of radiation damage. Slight attenuation of the amplitude seen in the spectra results from a low frequency quadrupole interaction, most probably due to some impurities or defects in the crystal structure. The experimental data were analyzed by using a model which included combined magnetic dipole and electric quadrupole interactions. The results show two distinct magnetic interactions at all temperatures in both cases. For Pd_2MnSn the characterstic Larmor frequencies measured at 10 K were found to be 319 Mrad/s for the component with major fraction and 119 Mrad/s for the minor fraction. The corresponding hyperfine fields (B_{hf}) are 21.2(1) T and 7.9(1) T, respectively. The higher frequency component was assigned to probe nuclei occupying Sn site and the lower frequency component to the Mn site in conformity with the previous results [3]. The PAC spectra for the Pd_2MnSb alloy were analyzed in a similar manner. The Larmor frequencies measured at 20 K are 350 Mrad/s (major fraction) and 320 Mrad/s (minor fraction). These were assigned to Sb and Mn sites, respectively, corresponding to hyperfine fields of 23.8(1) T and 21.3(1) T. The values of the hyperfine fields are in good agreement with those obtained in the previous studies [1, 2]. The temperature dependence of hyperfine fields is shown in Figure 2. After an additional annealing of the samples at 800°C for

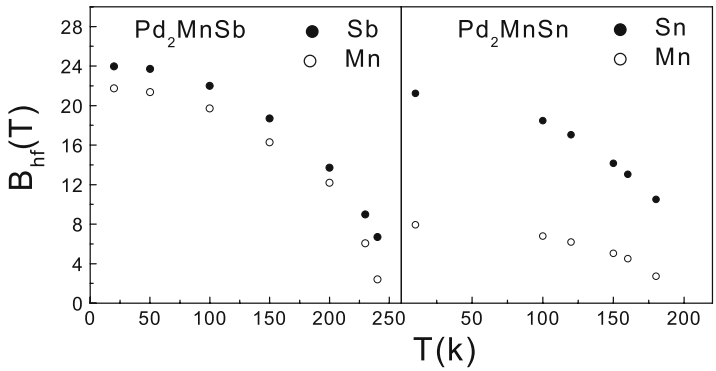

Figure 2. Temperature dependence of magnetic hyperfine fields in Pd_2MnSb and Pd_2MnSn as a function of temperature.

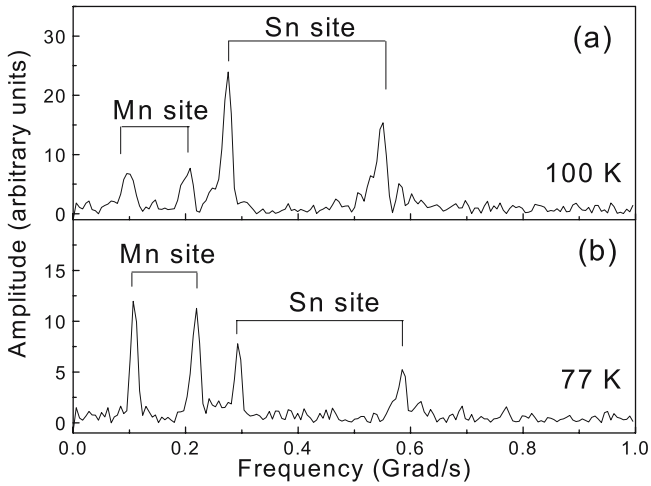

Figure 3. Fast Foureir spectra for the alloy Pd_2MnSn before (a) and after (b) annealing at 800°C for 24 h.

24 h in argon, the site occupation showed significant increase in the fraction of Mn site. This can be seen in the fast Fourier spectra before and after annealing at 800°C of Pd2MnSn (Figure 3).

We have now initiated a more systematic study of the site dependence of the hyperfine fields in the Heusler alloys Pd_2MnZ(Z = Sn, Ge, Sb) with the PAC technique. First measurements on the ternary alloys Pd_2MnSn and Pd_2MnSb with ^{111}In probes showed two Larmor frequencies corresponding to the Mn and Sn sites. This is already an interesting result as it was possible to simultaneously measure the hyperfine fields at more than one site in the same implanted sample. The reason why the implanted ^{111}In nuclei show little preference for Pd sites is not clear at the moment. It is possible, however, that the probe does enter the Pd site during implantation but migrates to other sites on annealing at lower tem-

peratures. Further measurements to better understand the annealing temperature dependence of the ^{111}In-site occupation as well as to obtain the hyperfine field at the Pd site if possible are planned. Present experiment has demonstrated that for samples where Pd is one of the components the process of implantation of ^{111}In using the present nuclear reaction is quite efficient compared to the conventional methods of introducing the probe in the sample. For the implantation of ^{111}In on samples that do not contain Pd the method will require some modifications. The ^{108}Pd(^{7}Li, 4n)^{111}In or ^{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.

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