Hyperfine Interactions **133:** 83–87, 2001.

PAC Measurements on New Ferromagnetic Compound Pd_2TiSn

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(Received: 26 June 2000)

Abstract. Magnetic hyperfine field (mhf) acting on ^{181}Ta at the Ti site has been investigated in the alloy Pd_2TiSn by Perturbed Angular Correlation (PAC) measurements using the (133–482) keV γ–γ cascade in ^{181}Ta following the β^- decay of ^{181}Hf. The magnetic hyperfine field was measured as a function of temperature in the range of 45–1000 K. The magnetization measurements were carried out with a vibrating sample magnetometer. The interpretation of the experimental results is based on the assumption that the probe nuclei occupy both the regular Ti sites as well as the Ti atom sites randomly distributed on the Pd sublattice.

Key words: magnetic hyperfine field, Heusler alloys, perturbed γ–γ angular correlation.

1. Introduction

A new family of intermetallic compounds of the type Pd_2TiZ, with Z = Al, In, Sn which order magnetically but do not contain any magnetic element was recently reported [1–3]. They are characterized by high Curie temperatures and relatively small magnetic moments. These inter-metallic compounds, with the stoichiometric composition X_2YZ in the ordered state, have the $L2_1$ structure type known as Heusler alloys [4]. The X element is usually a transition or noble metal such as Cu, Pd, Ni, Co; Y is a transition element such as Ti, Zr, Hf, V, Nb and Z is an sp element belonging from group IIIA to VA. The Heusler alloys usually exhibit magnetic behavior and their study [5] has made a unique contribution towards the understanding of the hyperfine field present at the impurity sites in ferromagnetic materials.

The alloy Pd_2TiSn was reported to have a relatively small magnetic moment per formula unit $\approx 0.005\mu_B$ [3] with a high Curie temperature. For the Pd_2TiAl alloy the magnetic moment per formula unit was measured to be $0.21\mu_B$ [2]. The Pd_2TiIn alloy was found to order antiferromagnetically below 110 K with a paramagnetic moment of $4.9\mu_B$, which is much higher than can be accounted for on the basis of a localized moment associated with the Ti atoms [1].

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In the present work the magnetic hyperfine field (mhf) has been measured on the ^{181}Ta probe at the Ti site in and Pd_2TiSn alloy using the perturbed γ–γ angular correlation technique. The experimental results are compared with the Co_2TiSn alloy and discussed in terms of the assumption that in the case of Pd_2TiSn the ^{181}Ta probe nuclei occupy two distinct sites, the regular Ti sites as well as the Ti atoms disordered on the Pd site.

2. Experimental

The alloy was prepared by repeated melting of the constituent elements with a purity of better than 99.9% under argon atmosphere in an arc furnace, with radioactive ^{181}Hf substituting about 0.1% of Ti atoms. The sample was annealed at 800°C during 24 h in argon atmosphere. The sample was analyzed by X-ray diffraction where it was verified that it had the expected $L2_1$ structure. The saturation magnetization measurements were made in a conventional Foner-type vibrating-sample magnetometer. The measurements were carried out in the temperature range of 295–460 K in an externally applied field of 0.1 T at which the magnetization was found to be saturated. The system was calibrated with a pure (99.999%) Ni foil.

The PAC measurements were performed with a conventional gamma spectrometer with a fast-slow coincidence setup using four BaF_2 detectors. The detector system had a time resolution of the order of 0.7 ns. The well-known (133–482) keV γ-cascade in ^{181}Ta populated by the β^- decay of ^{181}Hf was used to measure the TDPAC spectra. The perturbation factor for an unpolarized ferromagnetic sample can be written (neglecting the A_{44} term) as:

$$A_{22}G_{22}(t) = A_{22}\big[0.2 + 0.4\cos(\omega_L t) + 0.4\cos 2(\omega_L t)\big], \tag{1}$$

where $\omega_L = \mu_N g H_{hf}/\hbar$ is the Larmor frequency. With the known g-factor of the 482 keV ($5/2^+$) state of ^{181}Ta as $g_{5/2} = 1.3(1)$ [6] it is possible to determine the ^{181}Ta hyperfine field (H_{Ta}) from the measured Larmor frequency. The $A_{22}G_{22}(t)$ measurements were performed at several temperatures in the range from 45 to 1000 K.

The sign of the field was determined by placing the sample in an external polarizing magnetic field of about 0.5 T applied perpendicular to the plane of the detectors and measuring the ratio $R(t)$ at a fixed angle. The quantity $R(t)$ is expressed in terms of Larmor frequency as follows (neglecting the A_{44} term)

$$R(t, \theta = 3\pi/4) = \frac{N\uparrow - N\downarrow}{N\uparrow + N\downarrow} \approx -\frac{3}{4}A_{22}\sin(2\omega_L t), \tag{2}$$

where $N\uparrow$ and $N\downarrow$ are the number of coincidences with the externally applied magnetic field in the up ($\uparrow$) and down ($\downarrow$) directions, respectively. The measurement of $R(t)$ was made at 200 K for the Pd_2TiSn alloy using a cold finger refrigerated by liquid nitrogen.

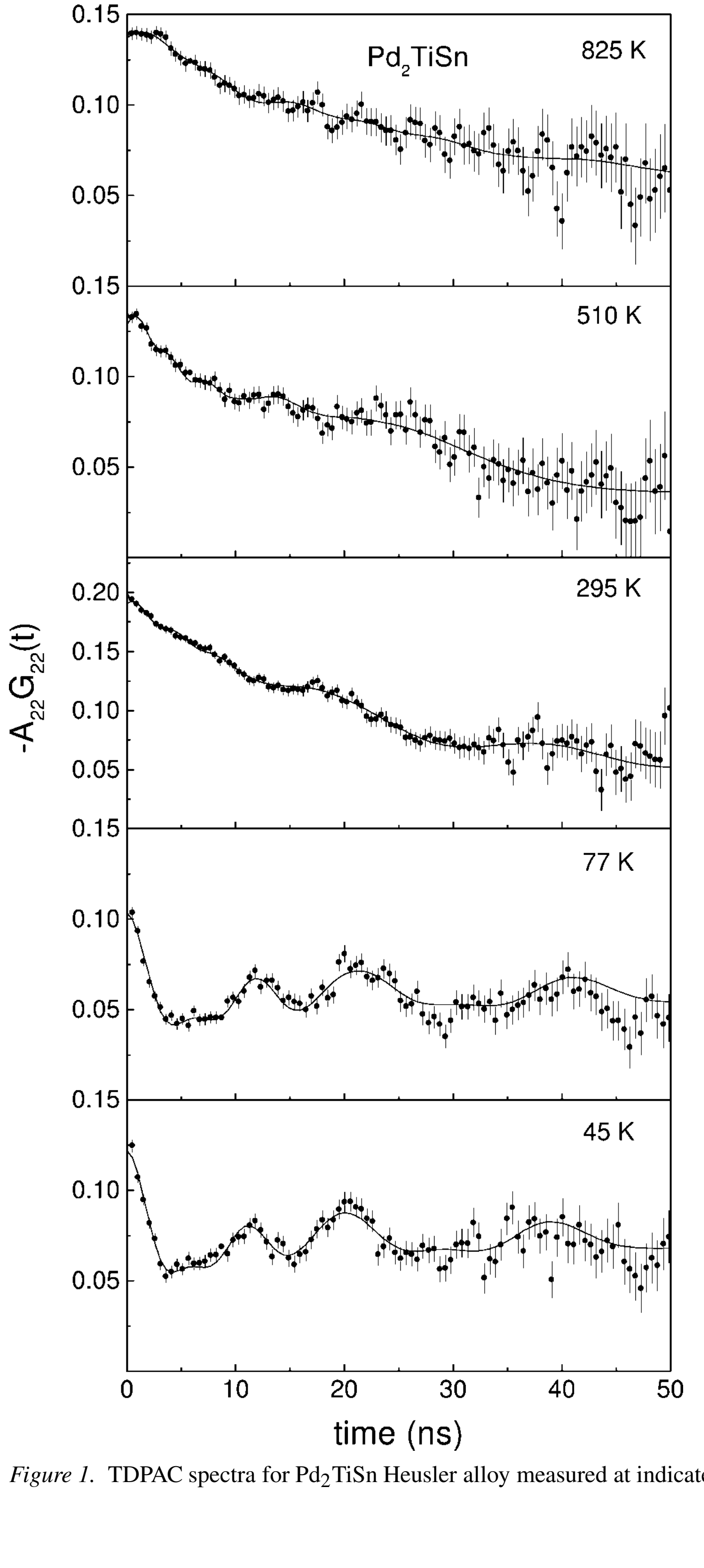

Figure 1. TDPAC spectra for Pd_2TiSn Heusler alloy measured at indicated temperatures.

3. Results and discussion

The value of the magnetic moment per formula unit determined from the magnetization measurements on the alloy Pd_2TiSn was found to be $0.037\mu_B$. Whereas this value of magnetic moment is still very small when compared with other Heusler alloys containing Co or Mn atoms, it is much larger than the previously measured value for Pd_2TiSn [3]. We do not quite understand the reason for this discrepancy.

Some of the PAC spectra obtained for the Pd_2TiSn alloy are shown in Figure 1. Solid curves are the least-squares fit of the experimental data to the expression (1). The results of PAC measurements show two distinct hyperfine fields with site populations of about 65% (H_1) and 35% (H_2), respectively. Relative population of the two field components is almost independent of temperature. Since the X-ray diffraction result did not reveal the presence of a second phase, we attribute this to ^{181}Ta probe atoms occupying two sites within the $L2_1$ structure. Figure 2 shows the parameter $R(t)$ as a function of time measured at 200 K. The solid curve is the least-squares fit of the experimental data to the expression (2). The result indicates a positive sign for the magnetic hyperfine field.

The H_{Ta} is plotted as a function of the temperature for the field components H_1 and H_2 in Pd_2TiSn in Figure 3. The component H_1 decreases quickly from about 8 T at 77 K to about 0.5 T at 300 K and then remains almost constant at this value. The component H_2 shows a similar behavior except that the initial drop in the value of H_2 between 77 K and 300 K is more gradual and at still higher temperatures the value stabilizes around 2 T.

The observed magnetic moment for the Pd_2TiSn alloy is much smaller compared to the value of $2.06\mu_B$ [7] per formula unit associated entirely with the Co atom in Co_2TiSn. The hyperfine field H_{Ta} at the Ti site in the Co_2TiSn alloy is

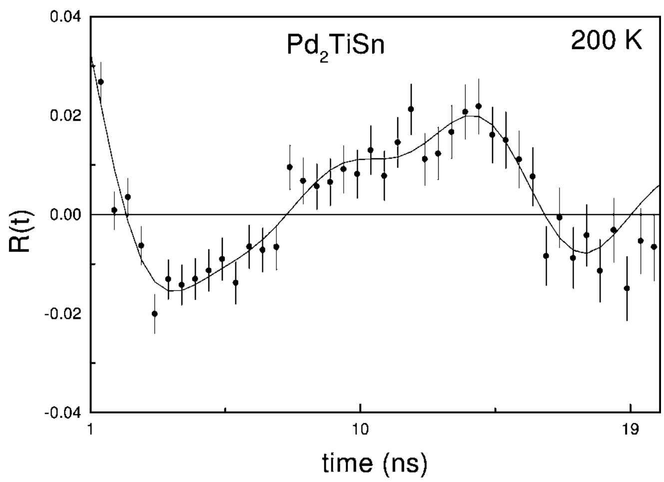

Figure 2. TDPAC spectrum for Pd_2TiSn Heusler alloy measured at 200 K with an external polarizing magnetic field.

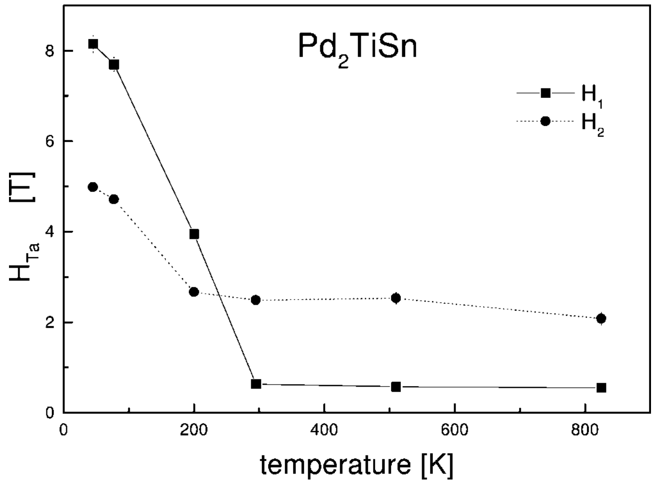

Figure 3. Temperature dependence of magnetic hyperfine field components H_1 and H_2 for Pd_2TiSn Heusler alloy.

known to be 48 T at 77 K and although the sign of the field was not measured it is believed to be negative from the known systematics of similar cobalt based Heusler alloys [5]. These observations contrast with the present results for the Pd_2TiSn alloy where the field is much smaller and its sign is positive. This indicates that the mechanism for the magnetic moment formation as well as the transfer of the spin density to the Ti atom site in the Pd-based Heusler alloys is quite different. Neumann *et al.* [3] have suggested the existence of two magnetic sublattices in the Pd_2TiSn alloy which account for its magnetic state. This seems to be in accordance with the two components of the magnetic hyperfine fields in this compound observed in the present study.

Acknowledgement

This work was partially supported by FAPESP.

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