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Lattice Site Dependence of a Cd Hyperfine Field in Pd_2MnSn Heusler Alloy

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Abstract. The magnetic hyperfine field B_{hf} for Cd in the Heusler alloy Pd_2MnSn at the site of chemically introduced Ag has been investigated by PAC following the β^- decay of ^{111}Ag. Sign and temperature dependence of B_{hf} have been determined. Comparison of the result $B_{hf}(T = 0) = +8.0(1)$ T with earlier data and *ab-initio* band structure calculations leads to the conclusion that the Ag activity has been incorporated at the Mn site.

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

1. Introduction

The intermetallic compounds known as Heusler alloys have stoichiometric composition X_2YZ and, in the ordered state, the $L2_1$ structure [1]. The X-element is usually a transition or noble metal such as Cu, Pd, Ni; Y is a transition element such as Ti, Zr, Hf, V, Nb and Z is an sp element belonging to group IIIA to VA. Most Heusler alloys are magnetic and their study has made unique contributions towards the understanding of the hyperfine fields B_{hf} present at the impurity sites in ferromagnetic metals. The Heusler alloys of interest here are of the form X_2MnZ. The local magnetic moment is carried by the Mn atoms and has values of about $4\mu_B$. In these alloys the sp element at the Z-site is the second nearest neighbour of the magnetic atom Mn, while the X-element is the nearest neighbour. Substitutional impurities on Mn sites will also have Mn atoms as 2nd nearest neighbours.

There is an appreciable quantity of experimental data on B_{hf} for substitutional impurities in Heusler alloys of the form X_2MnZ [2, 3], measured by different probes in a same host alloy as well as measurements using the same probe in different alloys. The great majority of these measurements are for the Z-site with only a few sp probe elements. It is interesting, however, also to study the magnetic hyperfine field acting on other sites. The Pd_2MnZ Heusler alloys are particularly interesting because depending on the Z-element they can exhibit ferromagnetism

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or antiferromagnetism. Pd_2MnIn is an antiferromagnetic alloy with the Mn spins positioned in such a way that the net magnetic field at the Z-site is equal to zero but at the Pd site it is not. The Pd_2MnSn alloy is ferromagnetic with Curie temperature of 189 K, a local magnetic moment of 4.3 μ_B and a lattice parameter of 6.38 Å [4].

The present work started as an attempt to determine the magnetic hyperfine field for Cd at the Pd site in Pd_2MnSn with radioactive ^{111}Ag incorporated by diffusion. The perturbed γ–γ angular correlation (PAC) technique was used to measure B_{hf} at ^{111}Cd and the results are compared to those previously measured [5, 6] with ^{111}In as parent for the same probe state at the Sn and (presumably) Mn site using PAC.

2. Experimental

The Heusler alloy sample of about 5 g was prepared by repeated melting of a mixture of the constituent elements in vacuum with an electron beam. It was cut with a diamond saw into slices about 0.3 mm thick. The ^{111}Ag activity was produced by irradiating ^{110}Pd samples of 0.5 mg with thermal neutrons in the BER2 reactor of the Hahn–Meitner Institut followed by chemical separation with ion exchange. The carrier-free ^{111}Ag solution was dropped on the surface of the polished samples that were then annealed for 24 h at 800°C in a sealed quartz tube under argon.

The time-differential perturbed angular correlation measurements on the 96–245 keV γ-cascade in ^{111}Cd were carried out with a conventional fast-slow coincidence setup using 4 NaI(Tl) detectors in square planar arrangement resulting in 12 time coincidence spectra. The perturbation factor for an unpolarized ferromagnetic sample consisting of randomly oriented domains can be written (neglecting the A_{44} terms) as:

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

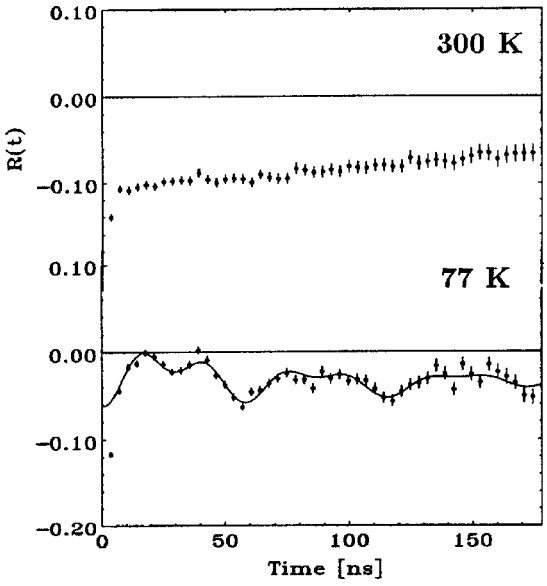
where $\omega_L = 2\pi g B_{hf}/h$ is the Larmor frequency. Using the accurately known g-factor of the 245 keV state in ^{111}Cd, $g_{5/2} = -0.305$ [7], the Cd hyperfine field is obtained from the measured ω_L. First experiments were performed without polarizing field at 77 and 300 K. Measurements at variable temperature were made using a helium refrigerator. The sample was placed in an external polarizing magnetic field of 0.2 T from a compact permanent magnet applied perpendicular to the plane of the 4 detectors. For this situation the ratio function $R(t)$ is expressed as:

$$R(t) = A_{22}\big[0.25 + 0.75\cos(2\omega_L t)\big]. \tag{2}$$

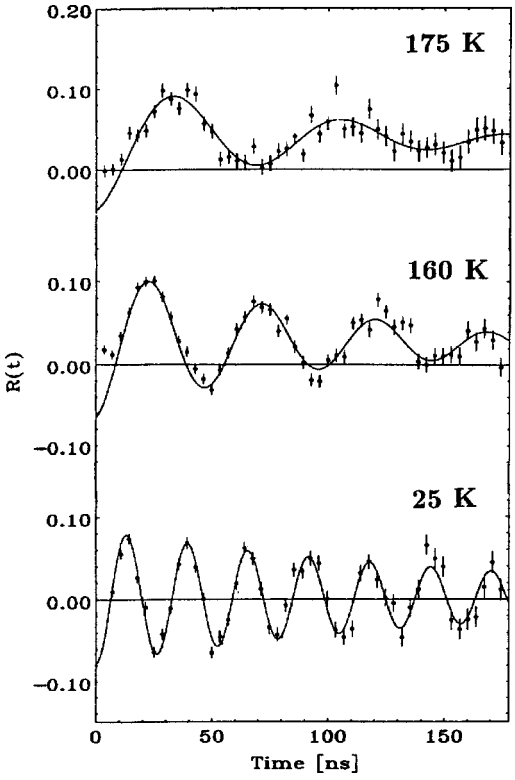
The sign of B_{hf} was obtained both from the effect of the external field on ω_L and by using detector angles of 45°.

3. Results

Results of the PAC measurements for the Heusler alloy Pd_2MnSn are shown in Figures 1(a) and (b). Solid curves are the least-squares-fit of the expressions (1)



(a)



(b)

Figure 1. (a) Zero-field PAC spectra. (b) PAC spectra in an external field.

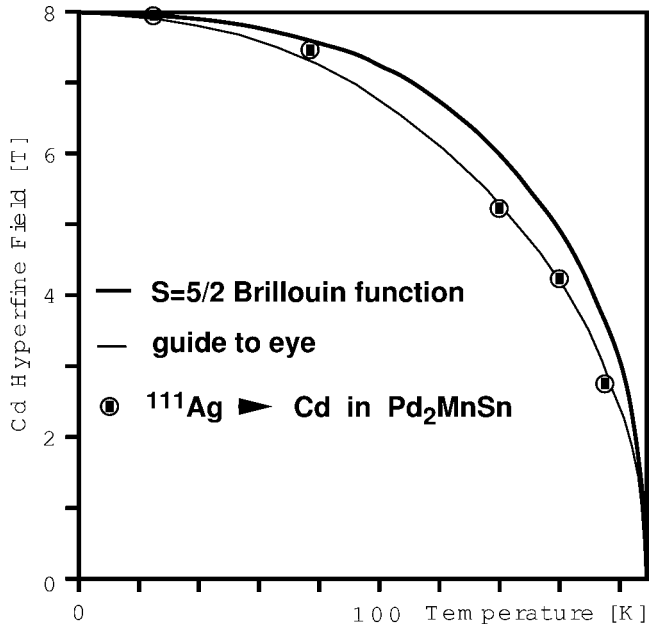


Figure 2. Temperature dependence of B_{hf}. The thick line is the $S = 5/2$ Brillouin function, the thin line a guide to the eye.

and (2), respectively, to the experimental data. The spectrum for room temperature shown in Figure 1(a) corresponds to the paramagnetic phase. At this temperature there is no magnetic interaction and, as Pd_2MnSn has cubic structure, the effective electric quadrupole interaction should be equal to zero for all substitutional sites. In agreement with this expectation, the data show only a weak interaction attributable to structural imperfections in the alloy. The frequency obtained from the spectrum at 77 K is $\omega_{\mathrm{L}} = 17.4 \pm 0.2$ MHz. Figure 1(b) shows representative spectra at various temperature below the Curie point taken with an external magnetic field. Figure 2 shows B_{hf} plotted as a function of measuring temperature. The value extrapolated to zero is $B_{\mathrm{hf}}(T = 0) = +8.0(1)$ T. It can be seen that the temperature dependence deviates somewhat from the magnetization curve expected from simple theory. In a previous work [5] B_{hf} for Cd on the Sn site in Pd_2MnSn alloy was measured by PAC using the 173–245 keV cascade from the ^{111}In parent. The magnetic hyperfine field reported was -20 T (extrapolated to 0 K), quite different from the present result.

4. Hyperfine field calculations

Modern methods based on density functional theory allow to calculate the magnetic properties of most solids with good precision. The bulk Heusler alloys and their hyperfine fields have been treated before. Making use of the supercell technique, recently employed with success for impurity hyperfine fields in Fe [8], we have

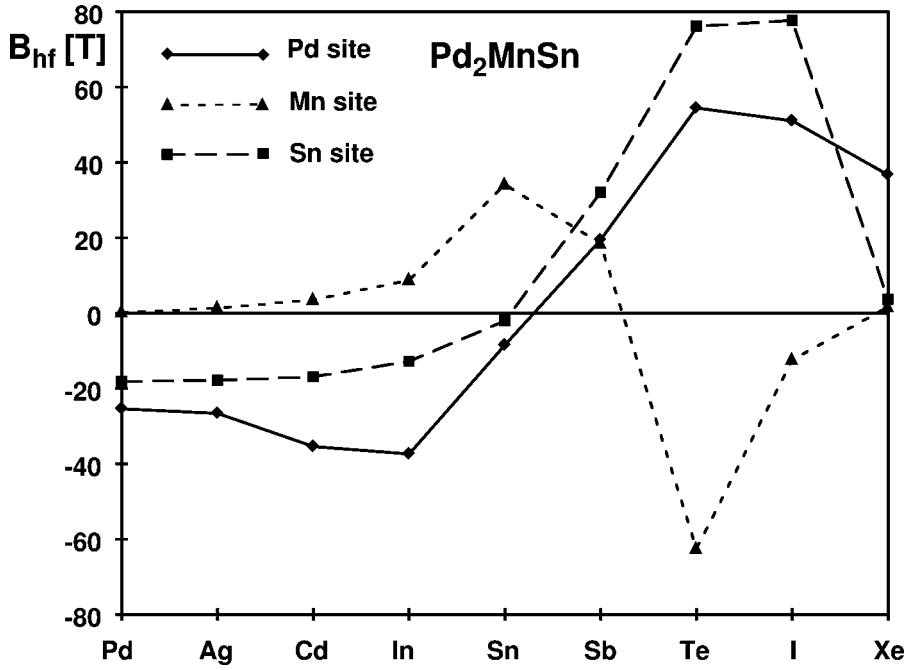


Figure 3. Calculated B_{hf} for 5sp impurities at the three lattice sites of Pd_2MnSn.

calculated the field acting on 5sp impurities at various sites in Pd_2MnSn with the computer code Wien97 [9]. Though the supercells used were with 16 atoms rather small and effects of lattice relaxation around the impurities were ignored, the results can serve as a guideline for the interpretation of the present experiment. As may be seen in Figure 3, fields for the early members of the series are large and negative at the Sn and Pd sites, while they are calculated to be small and positive at the Mn site. For the later 4sp elements a sign change and much larger values are predicted. This is fully in accord with the few data points available and suggests the assignment of the observed field value to Cd at the Mn site, in agreement with an earlier suggestion [6].

5. Conclusions

Ag introduced by diffusion into Pd_2MnSn is found to take up the Mn site under the present conditions. The sign and magnitude of the observed B_{hf} for Cd at the Mn site is reproduced by FLAPW calculations. The large and negative field calculated at the Pd site could perhaps be checked by repeating the present experiment using an alloy with controlled off-stoichiometry or by production of ^{111}Ag directly at the Pd site following *in situ* $^{110}Pd(n, \gamma)$ neutron capture.

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